Drop Coalescence in Liquid-Liquid Fluidized Beds

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A drop size segregation technique leads to a direct measurement of the interdrop coalescence frequencies of equisized droplet swarms in liquid-liquid fluidized beds. Mass transfer between the phases and the use of a tracer dye are avoided. The study includes the effects of highly viscous continuous as well as dispersed phases. Binary liquid solutions of ethylene glycol and distilled water provided aqueous phases of increasing viscosity, and toluene was the second liquid phase. Twenty two liquid-liquid systems including other dispersed liquids, namely, benzene, ethyl acetate, cyclohexane, and carbon tetrachloride, were studied by this technique.

In the case of toluene continuous, the drops of water dispersed in toluene were large in size, and they coalesced at unusually high rates up to about 100 times faster than those of toluene dispersed in water. Similarly, anhydrous ethylene glycol dispersed in toluene showed somewhat high coalescence frequencies. All the data excluding these two anomalous systems were analyzed by a stepwise regression technique to yield the best fitting correlation in terms of the most significant dimensionless ratios.

For the present, the proposed empirical correlation for the dimensionless parameter $[\omega D \times 10^5/V_s]$ containing the coalescence frequency (ω, time^{-1}) as a function of the coalescence number ($\sigma/\mu_c V_s$) and the viscosity ratio (μ_d/μ_c) is as follows:

$$(\omega D \times 10^5/V_s) = 0.020\,(\sigma/\mu_c V_s)^{1.18}\,(\mu_d/\mu_c)^{0.450}$$

SCOPE

The phenomenon of coalescence affects the interfacial area, the driving force, and hence the transfer coefficient of a given phase-contacting device. Coalescence followed by redispersion of the drops are likely to be responsible for the enhanced mass-transfer rates observed in agitated vessels (Mok and Treybal, 1971; Schindler and Treybal, 1968). Since coalescence alone of the dispersed phase diminishes the overall interfacial area, this phenomenon is detrimental in equipment like spray towers and direct contact heat exchangers. A correlation capable of predicting the rate with which a liquid-liquid dispersion will separate will help design an efficient settler.

Many workers have attempted to investigate this phenomenon by various approaches, namely, drop coalescence at a flat, stationary, liquid interface (Gillespie and Rideal, 1956; Charles and Mason, 1960; Lang and Wilke, 1971) and coalescence in agitated dispersions (Madden and Damerell, 1962; Curl, 1963; Groothius and Zuiderweg, 1964; Howarth, 1964, 1967; Coulaloglou and Tavlarides, 1976). All these methods employed some kind of a tracer dye or an indirect means in order to measure the coalescence frequencies and interpreted the nature of the drop-drop coalescence process indirectly. The experimental difficulty to accomplish a controlled head-on collision of two equisized drops forbade an early elucidation of this complex phenomenon.

The technique of a liquid-liquid fluidized bed surmounts all these difficulties and renders a simple means of visual observation and direct measurement of the coalescence frequencies in swarms of equisized drops in the absence of mass transfer between phases and impurities or a tracer dye.

The previous investigation (Maraschino, 1969) considered six organic liquids dispersed by water. The present study (Ramamoorthy, 1977) extends the number of liquid-liquid systems having a wide range of physical properties and includes the effects of highly viscous phases, both continuous and dispersed as well.

CONCLUSIONS AND SIGNIFICANCE

Liquids with widely varying physical properties were employed for the study of drop coalescence by the method of liquid-liquid fluidized beds (Ramamoorthy, 1977). The results include the effects of highly viscous phases, both continuous and dispersed. Binary liquid solutions of various compositions of pure ethylene glycol and distilled water provided aqueous phases of increasing viscosity; toluene was the second liquid phase. The addition of ethylene glycol to water increased also the solubility of the oil phase resulting in small drop diameters that varied by 2.2 fold.

During the study of toluene continuous, the behavior of water dispersed in toluene was anomalous because of the unusually high coalescence rates, large drop size, and large holdup in the bed. Anhydrous ethylene glycol also

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showed somewhat high coalescence rates. All the data except these two systems were employed in the regression analysis.

A stepwise regression technique (Draper and Smith, 1966) chose the most significant dimensionless ratios and yielded an empirical expression of maximum multiple correlation coefficient as follows:

$$(\omega D \times 10^5/V_s) = 0.020\,(\sigma/\mu_c V_s)^{1.18}\,(\mu_d/\mu_c)^{0.45}$$

where ω = coalescence frequency, $(\sigma/\mu_c V_s)$ = coalescence number, and (μ_d/μ_c) = viscosity ratio. The range of physical properties was as follows: $\rho_c = 1.0887 \times 10^3$ to 0.8603×10^3 kg/m^3, $\rho_d = 1.5901 \times 10^3$ to 0.7765×10^3 kg/m^3, $\mu_c = 5.976 \times 10^{-3}$ to 0.531×10^{-3} N s/m^2, $\mu_d = 18.63 \times 10^{-3}$ to 0.449×10^{-3} N s/m^2, $\sigma = 49.29 \times 10^{-3}$ to 5.7×10^{-3} N/m. Other variables ranged as follows: dispersed phase fraction = 15.4 to 1.5%, drop diameter = 0.450×10^{-2} to 0.205×10^{-2} m, coalescence frequency = 855×10^{-5} to 0.372×10^{-5} s^{-1}.

The coalescence behavior of polyphase dispersions is a subject of much investigation. Chemical engineers are concerned with the role of coalescence of the dispersed phase in mass transfer and separation operations. The tendency of a dispersed phase in the absence of stabilizers is to coalesce and separate in order to minimize its surface-to-volume ratio. In general, coalescence results in phase separation. Our ability to predict the rate of separation of a dispersion from the knowledge of the physical properties of the individual liquid phases will be most useful in the design of efficient liquid-liquid operations.

Coalescence is accompanied by a diminution of overall surface area, and it therefore undermines the transfer rate in spray towers, direct contact heat exchangers, and other mass transfer devices. In liquid extraction, the mixer settler unit poses diametrically opposite demands, namely, the avoidance of phase separation through coalescence by stirring in the mixer and the promotion of phase separation through coalescence by some sort of aids in the settler. Numerous data on various coalescing aids such as fiber beds, electric field, and vibrations are available in the literature. In the mixer, a dynamic equilibrium between coalescence and redispersion phenomena is presumed to exist. Therefore, an allowance for the coalescence phenomenon is incorporated in the prediction of drop size in agitated dispersions as a function of fractional holdup of the dispersed phase (Delichatsios and Probstein, 1976). A balance of the interfacial energy associated with the interfacial area before and after the coalescence of two equisized drops into a single, shuddering drop indicates that an excess interfacial energy associated with the new interfacial area is to be dissipated vigorously by viscous damping of the drop fluid. This violent action of coalescence helps establish a fresh gradient of the driving force (concentration or temperature) at the drop interface resulting in increased transfer rates. Enhanced mass transfer rates in agitated vessels have been observed (Mok and Treybal, 1971; Schindler and Treybal, 1968).

Successful coalescence of two equisized drops in another liquid by head-on collision has not been accomplished yet. Coalescence patterns were not consistent when two drops were launched from opposite nozzles (Scheele and Leng, 1971; Park and Crosby, 1965). The complexity of the phenomenon forced such investigations to a few simple situations. A great many data on the film drainage time or the rest time of a drop at a flat, stationary, liquid interface are available in the literature. Many correlations have been proposed for the drop rest time as a function of some dimensionless numbers (Davies et al., 1971). Some have attempted to extend their drop interface results to predict the drop-to-drop coalescence behavior and presented expressions for the coalescence frequency in the settling tanks. Many mechanisms for the drainage of the intervening liquid film have been developed (Gillespie and Rideal, 1956; Charles and Mason, 1960; Jeffreys and Hawksley, 1965; Lang and Wilke, 1971; Murdoch and Leng, 1971; Maraschino and Treybal, 1971). These developments offer little help in the prediction of the coalescence behavior of droplet swarms.

Others have used agitated tanks to study the coalescence of droplets (Madden and Damerell, 1962; Curl, 1963; Groothius and Zuiderweg, 1964; Howarth, 1964, 1967; Coulaloglou and Tavlarides, 1976). All such investigations, in some fashion, employed an indirect means such as a chemical reaction or mass transfer, a tracer dye, a step change in the agitator speed, or liquids of different densities, thereby restricting the interpretation of the general nature of the interdrop coalescence in droplet dispersions.

The technique of drop size segregation in the liquid-liquid fluidized bed surmounts all the foregoing difficulties. The method enables one to measure the coalescence fre-

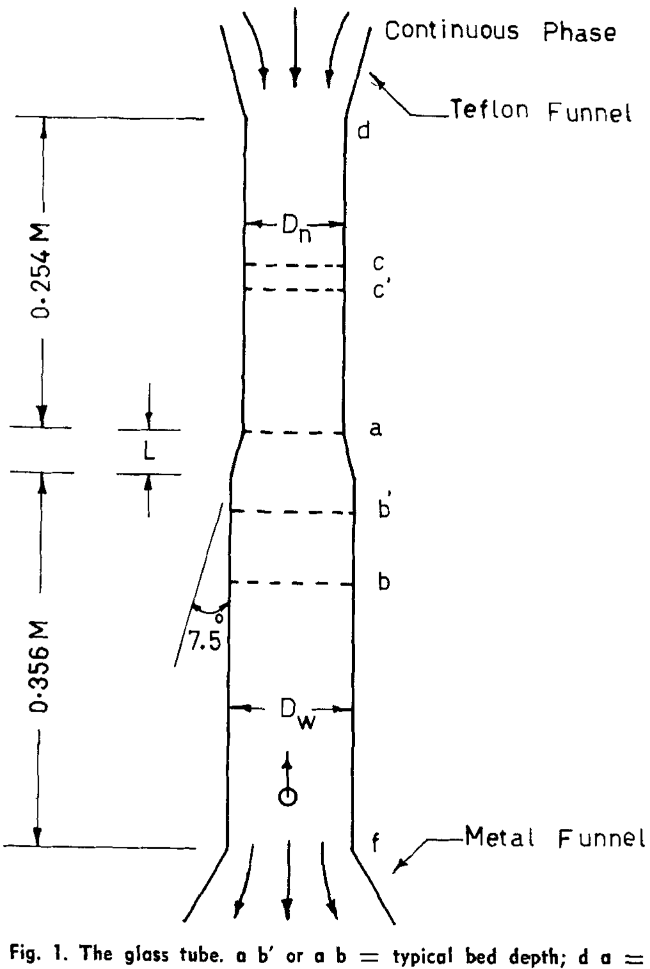

Fig. 1. The glass tube. a b' or a b = typical bed depth; d a = narrow section; b' f = wide section; c c' = observation section.

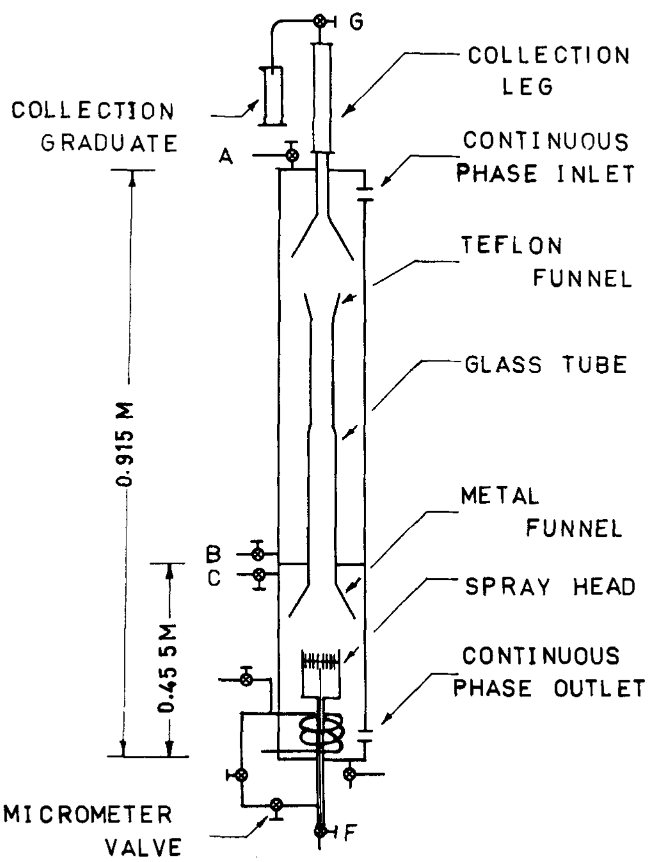

Fig. 2. Schematic diagram of the tank. A, B, C, F, and G denote regulating valves.

quencies in a swarm of uniformly sized drops by direct count. The coalesced drop leaves the bed immediately. The use of mutually saturated liquids of pure grade eliminated the mass transfer and the influence of contaminants. The liquids were in contact with stainless steel, brass, copper, glass, and Teflon only. Unhindered visibility of the tower allowed direct visual observation and precise control of the fluidized bed.

APPARATUS AND PROCEDURE

The apparatus is essentially a spray tower containing a specially designed glass tube to segregate the drops. Figure 1 shows one of the two glass tubes employed in this work. This glass tube was formed by joining two glass tubes of different diameters with a taper (Maraschino and Treybal, 1971). The continuous phase enters the narrow section and leaves from the wide section of the glass tube. Small, uniformly sized drops of the dispersed phase leave the nozzles of a spray head and enter the wide section. The flow rate of the continuous phase can be so adjusted that the small drops just barely fail to escape out of the tapered section. At this critical filter condition, the entering drops will accumulate in and below the taper to form a liquid-liquid fluidized bed of drops. By carefully adjusting the flow of the continuous phase, a well-behaved fluidized bed could be achieved. For a given drop size and glass tube, the fractional holdup in the liquid-liquid fluidized bed was fixed. If two drops that are moving randomly in the bed happen to collide and successfully coalesce to form a single large drop, and if this large drop which rises or falls faster will escape the bed immediately, it will pass through the narrow section and accumulate in the collecting tube.

Figure 2 shows the glass tube mounted inside the stainless steel tank with glass windows at the front and back; these windows could be easily removed for the purpose of cleaning and changing the glass tube. This tower as shown in Figure 2 was used to study the light dispersed phases; it was inverted for the study of the heavy dispersed phases. The drops issued from a spray head consisting of twenty four chamferred nozzles of stainless steel tubing 0.254 mm ID, 0.508 mm OD. Two glass tubes were of the following dimensions: first $D_n = 54.7$ mm, $D_w = 64.9$ mm, $L = 38.0$ mm (tube A); second $D_n = 59.8$ mm, $D_w = 65.3$ mm, $L = 22.6$ mm. Two different (D_n/D_w) ratios were employed to achieve correspondingly two different dispersed fractions for a given liquid pair and a given nozzle opening. An orificemeter, calibrated for the highly viscous continuous phases, was used to measure the flow rate during the experiment.

A constant temperature was maintained in the tower throughout the experiment. Before each experiment, the drop diameter was measured at the approximate conditions of actual flow rates of both the continuous and dispersed phases by collecting a certain volume of a known number of drops. Unlike the previous study (Maraschino, 1969), the batch method of studying the drop coalescence was not employed in the present work. Although the coalescence rates were very low in a majority of the systems, drops of the dispersed phase were continuously but very slowly added in order to maintain a constant depth of the fluidized bed. Though the coalescence frequency (as defined later) is not dependent upon the bed depth, a constant depth was necessary to obtain a reproducible values for the dispersed phase fraction and to avoid transient changes in the measurement of the coalescence frequency. Further details of the procedure are available elsewhere (Ramamoorthy, 1977). Figure 3 shows a typical liquid-liquid fluidized bed obtained for ethyl acetate dispersed in water.

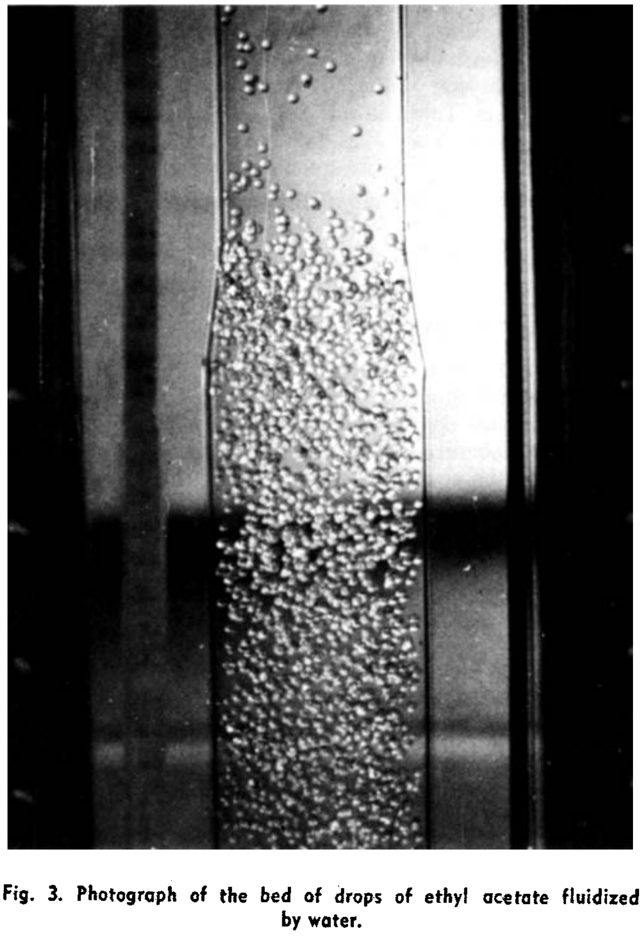

Fig. 3. Photograph of the bed of drops of ethyl acetate fluidized by water.

TABLE 1. SYSTEM IDENTIFICATION

No.	Dispersed phase	Continuous phase
1.	Benzene	Water
2.	Toluene	Water
	Viscous continuous phase	
3.	Toluene	66.6% Aqueous ethylene glycol
4.	Toluene	59.6% Aqueous ethylene glycol
5.	Toluene	51.4% Aqueous ethylene glycol
6.	Toluene	42.2% Aqueous ethylene glycol
7.	Toluene	33.25% Aqueous ethylene glycol
8.	Toluene	25.25% Aqueous ethylene glycol
9.	Toluene	16.6% Aqueous ethylene glycol
10.	Toluene	9.75% Aqueous ethylene glycol
11.	Toluene	2% Aqueous ethylene glycol
12.	Ethyl acetate	Water
13.	Cyclohexane	Water
14.	Carbon tetrachloride	Water
	Viscous dispersed phase	
15.	Water	Toluene
16.	14.5% Aqueous ethylene glycol	Toluene
17.	35% Aqueous ethylene glycol	Toluene
18.	49% Aqueous ethylene glycol	Toluene
19.	60.7% Aqueous ethylene glycol	Toluene
20.	69% Aqueous ethylene glycol	Toluene
21.	78% Aqueous ethylene glycol	Toluene
	Organic liquid-organic liquid	
22.	Anhydrous ethylene glycol	Toluene

Table 1 lists all the twenty two liquid-liquid systems that were studied by this technique. Table 2 lists the physical properties of these systems, the experimental drop sizes, and the dispersed phase fractions. The viscosities of aqueous ethylene glycol solutions were measured at a constant temperature to obtain a viscosity-composition curve which was used later to prepare aqueous phases of varying viscosities for the runs. The compositions listed in Table 1 are percent by volume of pure ethylene glycol. Care was taken to eliminate the hygroscopic absorption of moisture by ethylene glycol throughout the study. The slip velocities and coalescence frequencies varied from 10.74×10^{-2} to 4.12×10^{-2} m/s and from 855×10^{-5} to 0.372×10^{-5} s^{-1}, respectively.

EXPERIMENTAL RESULTS

In majority of the systems, only binary coalescence of two equisized drops was observed. In the case of some highly viscous continuous phases, ternary and multiple coalescence occurred. The coalescence frequency is defined in terms of the coalesced drops leaving the bed as

$$\omega = 2\ (\text{drop leaving rate})/N \qquad (1)$$

This definition is valid even for the case of multiple coalescence, since the process has to start with a binary coalescence in any event. Steady state was allowed to develop before each measurement of coalescence frequency. At these conditions, the dispersed phase fractions were reproducible. Only a fraction of the drop collisions in the fluidized bed result in coalescence. Howarth (1964) showed that the fraction of such successful collisions of equisized spherical particles in a turbulent field is proportional to $\omega\ D/(\overline{u^2(t)}\ \phi_d)^{0.5}$. Taking the slip velocity of the fluidized bed V_s as proportional to the root mean square of the fluctuating velocity component $(\overline{u^2(t)})^{0.5}$, the group becomes $(\omega\ D/V_s)/\phi_d^{0.5}$, where

$$V_s = \frac{V_c}{(1-\phi_d)} + \frac{V_d}{\phi_d} \qquad (2)$$

This dimensionless group ($\omega\ D/V_s$) involves three aspects, namely, the geometry, a characteristic of flow, and the coalescence phenomenon. Drop size and terminal velocity of rise or fall and hence the slip velocity of the fluidized bed are directly proportional to each other up to a certain critical size of the drop. Thus the net effect of their ratio (D/V_s) upon ω was expected to be minimal. The choice of V_s in the dimensionless group was therefore more than just convenient.

Owing to the lack of a sound theoretical model of the coalescence phenomenon, a dimensional analysis was attempted to arrive at some relevant dimensionless ratios that could best correlate the data. These ratios, namely, Re, We, Co, μ_d/μ_c, ϕ_d, and $\Delta\rho/\rho_c$ were employed to test the data individually obtained for each glass tube and overall data as follows:

$$\omega\ D/V_s = f(Re, Co, \phi_d, \mu_d/\mu_c, \Delta\rho/\rho_c) \qquad (3)$$

and

$$\omega\ D/V_s = f(Re, We, \phi_d, \mu_d/\mu_c, \Delta\rho/\rho_c) \qquad (4)$$

The parameter on the left-hand side of Equations (3) and (4) was expressed in the following dimensionless forms: $\omega\ D/V_s$, $\omega\ D/V_s\phi_d^{0.5}$, $\omega\ D/(\sigma/\mu_c)$, and $\omega\ D/(\sigma/\mu_c)\phi_d^{0.5}$. Even though some degree of arbitrariness was employed in obtaining various forms of this dimensionless parameter, the dimensionless ratios such as Re, We, and Co were treated as dimensionless ratios of forces and not merely an arbitrary grouping of variables. For example, the coalescence number, a ratio of interfacial force to viscous force, was found to be influential in describing drop coalescence in liquid medium, whereas the Weber number appears to effectively describe the drop coalescence behavior in air.

A stepwise regression analysis (Draper and Smith, 1966) of the data of each glass tube and of overall data exclud-

Table 2. Group Average Properties and Results

(1) No.	(2) ρ_c (kg/m^3)	(3) ρ_d (kg/m^3)	(4) $\mu_c \times 10^3$ (Ns/m^2)	(5) $\mu_d \times 10^3$ (Ns/m^2)	(6) $\sigma \times 10^3$ (N/m)	(7) $D \times 10^2$ (m)	(8) ϕ_d (%)
1A	997.9	876.3	0.958	0.626	32.05	0.343	8.27
1B	997.2	873.9	0.913	0.607	31.98	0.343	4.53
2A	997.7	864.6	0.995	0.578	34.61	0.342	8.11
2B	997.3	863.4	0.995	0.569	34.55	0.340	4.13
3A	1 088.5	868.0	5.944	0.561	16.07	0.219	2.38
3B	1 088.7	868.1	5.976	0.563	16.13	0.219	2.53
4A	1 081.6	868.8	4.946	0.476	16.08	0.239	2.00
4B	1 081.8	869.1	5.007	0.478	16.20	0.238	2.96
5A	1 072.1	868.3	3.843	0.511	16.29	0.242	5.11
6A	1 060.6	869.2	3.060	0.559	20.60	0.263	5.64
6B	1 060.6	869.3	3.066	0.560	20.63	0.263	2.45
7A	1 047.5	867.1	2.255	0.549	21.28	0.268	5.37
7B	1 048.4	868.8	2.375	0.560	21.30	0.268	3.63
8A	1 038.2	869.2	1.987	0.633	23.05	0.294	7.75
8B	1 037.8	868.3	1.933	0.616	23.00	0.290	3.68
9A	1025.2	868.7	1.521	0.552	27.13	0.310	7.77
9B	1 025.4	869.2	1.543	0.555	27.18	0.310	4.02
10A	1 014.1	869.0	1.268	0.583	30.10	0.321	8.21
10B	1 014.0	868.8	1.261	0.582	30.08	0.321	4.01
11A	1 000.5	865.2	1.055	0.565	34.69	0.335	8.64
11B	1 000.1	864.0	1.022	0.552	33.75	0.335	4.08
12A	996.9	898.2	1.072	0.449	5.70	0.205	2.01
13A	998.6	777.4	0.999	0.918	46.81	0.324	10.90
13B	998.3	777.7	0.976	0.902	46.76	0.337	4.34
14A	997.9	1 590.1	0.995	0.974	49.29	0.221	4.49
15A	861.6	996.8	0.555	0.994	34.47	0.450	15.4
15B	860.3	996.3	0.544	0.993	34.40	0.450	7.89
16A	862.1	1 016.6	0.531	1.328	30.83	0.413	9.88
16B	863.0	1 016.9	0.537	1.361	30.87	0.413	7.42
17A	863.6	1 045.5	0.548	2.357	23.66	0.343	8.81
17B	864.1	1 045.7	0.551	2.394	23.72	0.343	4.63
18A	864.9	1 064.5	0.549	3.668	19.77	0.303	8.29
18B	863.6	1 063.8	0.540	3.517	19.59	0.312	3.70
19A	864.1	1 078.8	0.546	4.956	17.01	0.290	7.11
19B	864.0	1 078.7	0.544	4.926	16.95	0.287	3.26
20A	864.0	1 084 9	0.551	6.292	16.07	0.283	6.42
20B	863.8	1 084.7	0.549	6.216	16.04	0.283	3.05
21A	862.9	1 091.6	0.636	7.843	14.05	0.262	4.47
21B	863.4	1 092.0	0.640	7.990	14.18	0.260	3.13
22A	865.2	1 105.8	0.557	18.629	9.78	0.228	2.15
22B	864.4	1 105.2	0.551	18.00	9.51	0.230	1.47

'A' refers to results of tube A and 'B' to those of tube B.

ing the two anomalous systems of water and anhydrous ethylene glycol as dispersed phases was carried out for all possible functional relationships (linear, semilogarithmic, and logarithmic) between the dependent and the independent variables. The analysis tested a set of 169 data points for the best fit by choosing stepwise those dimensionless ratios that maximized the multiple correlation coefficient. In general, the dimensionless parameter D/V_s was found to correlate best with the data. The viscosity ratio showed a strong influence on nearly all the possible relationships. The R^2 values, the partial F values, and the variance for each of the fifty possible models were examined. The following correlation was found to fit the data with $R^2 = 0.86$:

$$(\omega D \times 10^5/V_s) = 0.020\ (\sigma/u_c V_s)^{1.18}\ (u_d/u_c)^{0.450} \tag{5}$$

Figure 4 shows the fit of the data with the proposed correlation.

DISCUSSION

Of those attempted, the proposed correlation contains the fewest number of dimensionless ratios and therefore only three empirical constants. No variables except V_s are repeated on the right-hand side of the correlation so as to eliminate the inadvertent smoothing effect. This correlation, unlike that of the previous work (Maraschino, 1969), covers a wide range of physical properties. Furthermore, it describes the coalescence behavior of an organic liquid either dispersed or continuous. The experimental data for toluene, dispersed as well as continuous, merged into a single trend by overlapping each other. It is not clear, at present, why pure water and pure ethylene

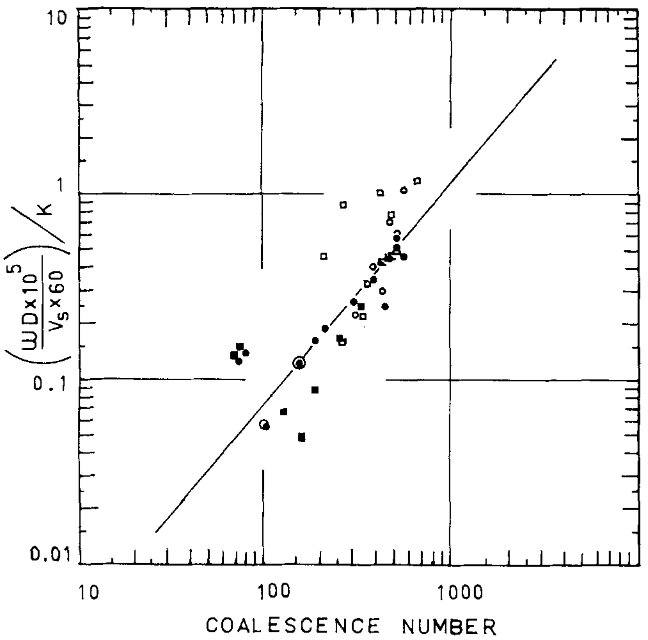

Fig. 4. Correlation of data. Circles = tube A; squares = tube B. Solid circles and squares = rising drops; blank circles and squares = falling drops.

glycol behaved anomalously while their solutions followed the general trend. These two results were therefore omitted from the analysis.

In spite of the wide range of liquid-liquid systems studied, the functional relationship of the dimensionless ratios was similar to the one obtained by Maraschino (1969). While all the combined data indicate the same functional dependence of the coalescence frequency, the magnitude of the coalescence frequency is however less certain. The present correlation fits the previous results of Maraschino (1969) with a constant of 0.40 instead of 0.020. First, it should be noted that the data fitting of a large range of variables is likely to differ from that of a small number of data. Second, the probabilistic nature of the coalescence process causes further discrepancies. Numerous data available on the coalescence times or the drop rest times for the coalescence of a drop at a flat, stationary, liquid interface show a distribution of times. For example, Charles and Mason (1960) measured the drop rest times of water drops at a benzene-water interface to have a range of coalescence times 0.20 to 14.0 s and an average time of 4.8 s. Gillespie and Rideal (1956) had measured the coalescence times of water drops at a benzene-water interface to be in the range 1.2 and 4.2 s for different diameters of water drops. In spite of the cleaned interface, the coalescence times followed a distribution owing to different patterns of film thinning that could occur for identical conditions of experiment conducted in identically the same apparatus (Liem and Woods, 1974). If such a scatter is observed in the measurements of the rest times of drops resting at a stationary, clean, liquid interface, it is likely to occur in the case of a fluidized bed of randomly moving drops. Keeping in mind the sensitive nature of the coalescence process, extreme care was taken to use only pure reagents in experiments and not to reuse any of them. With all these precautions, data scattering could not be reduced much further.

Drop Size

Since only one nozzle diameter was used throughout this investigation, the drop diameters varied by 2.2 fold owing merely to changes in the physical properties of the liquids. In most of the cases, the observed diameter of a drop agreed well within 5% of the value given by the Scheele-Meister correlation (Scheele and Meister, 1968). But, in the case of water and aqueous glycol dispersed, the drop sizes of the dispersed phase disagreed with the Scheele-Meister prediction by at least 20%. The size of water drops was very much larger. It must be pointed out that Scheele and Meister did not disperse water nor an aqueous phase in oil phase, and they formed drops in a stationary continuous phase. The aqueous phases or water did not wet the nozzle tip because the nozzles were chamferred.

Holdup

For a given liquid-liquid system and the glass tube combination, the fractional holdup is uniquely fixed (Treybal, 1963). Values of fractional dispersed phase holdup obtained for the twenty two liquid-liquid systems and for each of the two glass tubes varied from 15.4 to 1.5%. The measurements were reproducible, but the values were often greater than those predicted by the available correlations (Treybal, 1963). In the case of water dispersed in toluene, the dispersed phase holdup was considerably larger than the case of toluene as the dispersed phase. Since the slip velocity is defined in terms of the fractional dispersed phase holdup, the effect of holdup is included as V_s in the proposed correlation.

Wake Effect

The flow of continuous phase into the wakes formed in the rear of the droplets of the dispersed phase in the fluidized bed is likely to influence the value of the slip velocity for a given liquid-liquid system. Owing to the lack of data on the influence of wakes for a large number of liquid-liquid systems (Yeheskel and Kehat, 1973), the slip velocity values are reported elsewhere without the correction due to wakes (Ramamoorthy, 1977).

Effects of Continuous Phase Viscosity

The coalescence time of a drop at a flat, stationary, liquid interface is presumed to be principally dependent upon the drainage time of the intervening fluid. Different thinning patterns have been observed, and various film thinning mechanisms have been proposed. Similar mechanisms are assumed to hold for the drop-to-drop coalescence. One can therefore expect the coalescence time to vary directly with the viscosity of the intervening fluid, in this case the continuous phase. The proposed correlation agrees with this hypothesis and shows a strong inverse relationship between the coalescence frequency and the continuous phase viscosity.

As ethylene glycol was added to water in order to increase the viscosity of the continuous phase, it also lowered the interfacial tension with toluene, the dispersed phase. Because of the low interfacial tension, the drops were small, but the drag exerted on these drops was large, in some cases giving rise to multiple coalescence. Therefore, there was a limit to which the viscosity could be increased in the aqueous ethylene glycol-toluene system.

Dispersed Phase Viscosity

The phenomenon of internal circulation of a drop liquid confined by a mobile interface is well known. The tangential stress at the interface is continuous from exterior to interior of the drop, and this stress increases as the viscosity of the drop liquid increases, thus reaching the rigid sphere behavior at infinite viscosity. Therefore, one can expect the intervening fluid to overcome greater resistance caused by a highly viscous drop liquid at the interface, and thus the drainage time or the coalescence time can be expected to vary inversely with the dispersed phase viscosity in general. However, for a given liquid-liquid system, the ratio of the viscosities is a measure of the relative motion of the two fluids near the interface *that is completely mobile.*

Other Effects

The natural tendency of a phase is to exist in a minimum surface-to-volume configuration, and this tendency is a result of the interfacial tension. The proposed correlation agrees with the idea that, in general, the coalescence time is inversely proportional to the interfacial tension. The interfacial tension is a major factor in determining the size of a drop formed slowly at a chamferred nozzle. The effect of drop diameter on coalescence frequency is thus strongly related through interfacial tension.

The slip velocity is determined by the holdup measurements. The dispersed phase holdup is a function of the terminal settling/rising velocity which is, in turn, another function of the drop diameter which is mostly determined by the interfacial tension. It is therefore difficult to pin down individual effects.

Aqueous Phase Dispersed

Nearly all the studies on drop coalescence were confined to the coalescence of water drops at an oil-water interface only. A noteworthy observation in the present work was the unusually fast coalescence frequencies (about 100 times) of water drops dispersed in toluene compared to those of toluene drops dispersed in water. This unusual behavior of water dispersed in hydrocarbons is commonly known in liquid-liquid operations but as yet no quantitative measure of the anomalous behavior has been reported. One would suspect that the small viscosity of toluene was the cause for the exceptionally fast coalescence behavior of water drops. It remains unexplained why the drops of various aqueous solutions of glycol did not similarly behave anomalously. Fast coalescence behavior was also observed in the case of anhydrous ethylene glycol, a waterlike substance. These observations suggest a possible role of chemical forces in coalescence phenomenon.

Further, earlier studies of water drop coalescence at a flat, liquid interface indicate that small amounts of surface active agents had little effect upon the coalescence rates. If this be true, greater agitator power may be needed to break the fast coalescing water drops dispersed in hydrocarbon in agitated tanks. Depending upon the geometry and the homogeneity of turbulence field in a given agitated tank, it remains to be seen whether a drop size distribution may yield a larger mean diameter for water dispersed in a nonpolar hydrocarbon liquid than that for the hydrocarbon dispersed in water.

NOTATION

Co = coalescence number $(\sigma/\mu_c V_s)$, dimensionless
D = diameter, L
K = $(\mu_d/\mu_c)^{0.450}$
L = length of taper, L
N = total number of drops in the fluidized bed dimensionless
R^2 = multiple correlation coefficient, dimensionless
Re = Reynolds number $(DV_s\,\rho_c/\mu_c)$, dimensionless
$(\overline{u^2(t)})^{0.5}$ = root mean square of the fluctuating velocity component, L/t
V = superficial velocity, L/t
We = Weber number $(DV_s^2\,\rho_c/\sigma)$, dimensionless
$\Delta\rho$ = density difference, M/L^3
μ = dynamic viscosity, M/Lt
ρ = density, M/L^3
σ = interfacial tension, M/t^2
ω = coalescence frequency, t^{-1}
ϕ = volume fraction, dimensionless

Subscripts

c = continuous phase
d = dispersed phase
n = narrow section of glass tube
s = slip velocity of the fluidized bed
w = wide section of glass tube

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A Comprehensive Experimental Investigation of Tubular Entry Flow of Viscoelastic Fluids.

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Part II. The Velocity Field in Stable Flow

Center-line velocities and developing velocity profiles are presented for viscoelastic fluids flowing in a 2:1 and 4:1 abrupt entry contraction. Two flow regimes are clearly identified: a vortex growth regime and a divergent flow regime. Dramatic differences in the velocity field are observed and discussed for the two flow regimes. A kinematic model based on the velocity field data is proposed for vortex growth flow.

VELOCITY PROFILE MEASUREMENT

In parallel with the determination of the vortex characteristic discussed in Part I, point velocity measurement were made in the region just upstream of the 4:1 and 2:1 contractions. Of interest was the development of the axial velocity at the tube center line and the rearrangement of the axial velocity profile across the tube radius in the entry flow region, for both the vortex growth and the divergent flow regimes.

The two test sections used in the work were designed with long calming lengths upstream of the contraction plane to allow for dissipation of upstream disturbances before the fluid entered the contraction. To make sure that no disturbances were present, the axial velocity profile well upstream of each contraction was measured and compared with the theoretical fully developed profile. The accuracy of point velocity measurements was also checked by numerically integrating the velocity-radial position profiles and comparing the result with the separately measured volumetric flow rate. In all cases, the experimental profiles well upstream agreed to with 5% of the theoretical fully developed profiles, and the volumetric flow rate found by profile integration was within ±2% of the independently measured fluid flow rate.

The experimental conditions at which velocity measurements were made are summarized in Table 1. The parameters are calculated at the wall shear rate for fully developed flow in the downstream tube. Cases V1.1, V3.1, and V3.2 are in the vortex growth region, whereas V1.2, V1.3, V3.3, and V3.4 are in the divergent flow regime (see Figures 7, 8, and 11 in Part I). In general, the presentation of results which follows will differentiate between these two regions in order to establish the kinematic characteristics of each.

CENTER-LINE VELOCITY PROFILES

The development of the axial velocity at the tube center line from the upstream fully developed region to the contraction plane is shown in Figure 1 for fluid V3 in the 4:1 geometry. The axial velocity is shown in dimensionless form with respect to the average upstream velocity. In the vortex growth regime (V3.1, V3.2), for $N'_{Re} < 20$ and $N_{WS} < 0.5$, the axial velocity

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