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Drop Interface Coalescence in Liquid-Liquid System

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Abstract

This investigation is a study of the length of time where drops can exist at an oil-water interface before coalescence takes place with a bulk of the same phase as the drops. Many factors affecting the time of coalescence were studied in this investigation which included: dispersed phase flow rate, continuous phase height, hole size in distributor, density difference between phases, and viscosity ratio of oil/water systems, employing three liquid/liquid systems; kerosene/water, gasoil/water, and hexane/water. Higher value of coalescence time was 8.26s at 0.7ml/s flow rate, 30cm height and 7mm diameter of hole for gas oil/water system, and lower value was 0.5s at 0.3ml/s flow rate, 10cm height and 3mm diameter of hole for hexane/water system. It is observed that time of coalescence increased with increase in the dispersed phase flow rate, continuous phase height, hole size in distributor, and viscosity ratio of oil/water system. The results have been analyzed by dimensional and statistical analysis, and a correlation was developed relating coalescence time with the operating factors and the physical properties of the three oil/water systems.

Keywords: liquid-liquid system, drop coalescence.

Introduction

Coalescence is associated with the decrease in free energy of the liquid-liquid interface and is aided by the suppression of turbulence, which helps the droplets to aggregate to form a heterogeneous dense packed zone at the main interface between the bulk phases. The rate of migration of droplets to the coalescing main interface depends on the type of dispersion and the properties and interfacial characteristics of the system. The actual coalescence mechanisms are complex, involving the factors that govern the thinning of the continuous phase film between the two coalescing interfaces; depending on conditions, coalescence may occur either at the plane interface or at the drop/drop interface. Coalescence at the plane interface occurs at the exit end of a contactor after mass transfer is over, whereas drop-drop coalescence arises both within the droplet band awaiting coalescence and within the contactor [1]. The system of forces acting on a drop during the coalescence process was generally extremely complicated. These forces may be divided into three categories [2]:

1. Driving forces: those pushing the droplets towards the other interface.
2. Repelling forces: those in the opposite direction, repelling the interfaces from each other.
3. Resisting forces: those hindering the flow of the film of continuous phase, generally the hydrodynamic viscosity.

Numerous studies of the coalescence of drops at plane interfaces had been made for two-component systems. These studies with flat interfaces had helped in the understanding of coalescence without the complexities of drop-drop and multidrop interactions. Investigators had found that the time interval between the arrival of a droplet at an interface and final coalescence is not constant but exhibits a distribution of time although the distribution is approximately Gaussian [3]. Furthermore, all workers accept that the coalescence processes takes place through five consecutive stages [4]:

1. The approach of the drop to the interface, resulting in deformation of both the drop and the interface.
2. Damped oscillation of the drop at the interface.

3. The formation of a film of the continuous phase between the drop and its bulk phase.
4. Drainage of the film, its rupture and removal with initiation of the coalescence process proper.
5. Transference of the contents of the drop (partially or wholly) into its bulk phase.

The mean coalescence time t_m, which is also termed the rest time, comprises the mean of several observations of the total time taken for stages 1 to 5. The time taken for stage 1 is termed the predrainage time and that for stage 2 and 3, the drainage time. The coalescence time denotes the sum of pre drainage and drainage times. The time taken for film removal (after its rupture) and deposition of drop contents had been found to be almost negligible (of the order of 0.06-0.08s).

Brown and Hanson [5] had shown that single-drop coalescence occurs in one or more steps. If the simplest possible case of a single aqueous drop coalescing at a plane oil-aqueous interface is considered, it is found that instantaneous single-staged coalescence rarely occurs. When it first reaches the interface, the drop is prevented from coalescing with the underlying aqueous phase by a thin film of oil trapped between its flattened undersurface and the interface. This film drains radially outwards under the gravitational force exerted by the drop until it is approximately 1μ in thickness, when it ruptures, with the subsequent drainage of the drop into its home-phase.

Charles and Mason [6], using high-speed photography, were able to show that during the drainage process the drop forms into a vertical column of liquid, the height of which remains virtually constant while the radius decreases. When the circumference of the liquid cylinder becomes equal to its height, it behaves as an unstable jet in which a Rayleigh type disturbance is concentrated at the base of liquid column can grow, when its amplitude becomes equal to the radius of column, a break occurs, the remaining undrained liquid forming a new droplet (the secondary droplet). This process may be repeated several times. The phenomenon was called *Step-wise coalescence*.

The coalescence of a liquid drop at a plane interface is controlled mainly by the following factors, which affect the draining and rupture of the continuous phase [7]; It has been postulated that the mean coalescence time has a proportional dependence on the drop size of the form (t_m α dn) and found n is 3 [6]. Lawson [8] had reported from his results that the power on d was 1.5, and an increase in droplet size increased the drainage force due to the weight of the drop, but the volume of continuous phase in the film also increased . Hanson and Kaye [9] had found that the settling distance of the drop to the interface affected the coalescence time. Jeffreys and Hawksley [10] had shown that coalescence time and the stability of the drop increased with increase in the distance of fall. Generally, it had been accepted that the coalescence time t was proportional to L^n where L was the distance of fall and the exponent n increased with drop size, but was independent on temperature. Jefferys et al. [11] reported the distance of fall of the drop could either increase or decrease the stability of the drop, depending on the thermal or mechanical disturbances produced. The large differences in density result in severe deformation of the drop. However, Lawson [8] confirmed that the coalescence time increased with increase in the density difference between the phases. Lang and Wilke [12] were one of many workers included this factor in models and correlations.

Jeffreys and Hawksley [10] correlated coalescence time with physical properties of the system by considering the factors which affect coalescence time in dimensional analysis, estimating the significance of the parameters and exponents by factorial experimentation. The resulting correlation was:

$$t_{1/2} = 4.53 \times 10^{5} \left[\left(\frac{\mu^{1/2} \Delta\rho^{1.2}}{\gamma^{2}} \right) \left(\frac{T}{25} \right)^{-0.7\mu^{1/2}} \times d^{0.02} \left(\frac{\gamma^{2}}{\mu^{1/2}} \right)^{0.55} L^{0.001} \left(\frac{\gamma^{2}}{\mu^{1/2}} \right) \right]^{0.91} \qquad (1)$$

where $t_{1/2}$ is the half-life rest time which is the time taken for half the drops in the sample studied to coalesce, generally $t_{1/2}$ has been found to be more reproducible than t. Jeffreys et al. (11) simplified the analysis by restricting their investigation, by stating that temperature affected the physical properties only so that temperature, as such, need not be considered as a variable. The resulting correlation became:

$$\frac{\gamma t}{\mu d} = 1.32 \times 10^{5} \left(\frac{L}{d} \right)^{0.18} \left(\frac{d^{2} \Delta\rho g}{\gamma} \right)^{0.32} \qquad (2)$$

This correlation has been modified by Smith and Davies [13] for use when neighboring drops influence the drainage rate of continuous phase film (as in the case of a dense drop buildup beneath the main interface of a spray column):

$$\frac{\gamma . t}{d_{o} \mu_{c}} = 31 \times 10^{3} \left(\frac{d_{o}^{2} \Delta\rho g}{\gamma} \right)^{-1.24} \left(\frac{\mu_{d}}{\mu_{c}} \right)^{1.03} \qquad (3)$$

Several techniques have been used for measuring and observing the shape, drop size, and interfacial area. Kintner et al. [14] presented high-speed motion picture techniques to permit observation of the nature and progress of such fast phenomena as drop formation, coalescence or breakup. For liquid drops moving in a liquid medium, which are more difficult to photograph due to the much smaller differences in densities and refractive indices, the simplest solution was to dye the drop to produce high contrast and a tan and light brown reflecting background.

Aziz [15] measured photographically drop shape and size using a Zenit camera with an extension tube. A shutter speed of 1/500 sec and aperture setting of f/5.6 was used the column wall distortion due to curvature was avoided by using a Perspex box filled with water which surrounded the column. Klinger [16] used the same technique to measure the drop sizes and investigate the phenomenon of drop coalescence and breakage in pulsed packed extraction column.

Recently, The Particle Image Velocimetry (PIV) method was employed to determine vertically planar velocity fields of drops surrounded by liquid ambient that underwent coalescence. In addition, high-speed video camera was able to track the extremely rapid thin film retraction following rupture and the interfacial deformation during drop fluid collapse into the underlying liquid (17). Kassim and Longmire (18) also used the Particle Image Velocimetry (PIV) method to determine instantaneous velocity and vorticity fields in vertical planes of flow and to identify interesting regions and events throughout the impact.

Experimental Work

Equipment Description

In this study, the following facilities were used (the general view of the equipment is shown in Fig. 1. Coalescence column is graduated QVF column of 8cm inner diameter and 40cm height was used to show the coalescence of droplets at oil-water interface. Distributor was of 9.8cm diameter placed at the bottom end of the column. The dispersed phase distributor at the bottom of the column was constructed of Perspex so that it was not wetted by the aqueous phase. There were five different distributor plates, each one contained three holes of 3, 5, 7, 9 and 11mm in diameter for each distributor, respectively. They were perforated as the triangular pitch and 2.5cm the distance between adjacent holes [19] shown in Fig. 2. Dosing pump used to draw the oil fluid from oil vessel and pass through the distributor at the end of the column to create a different flow rate.

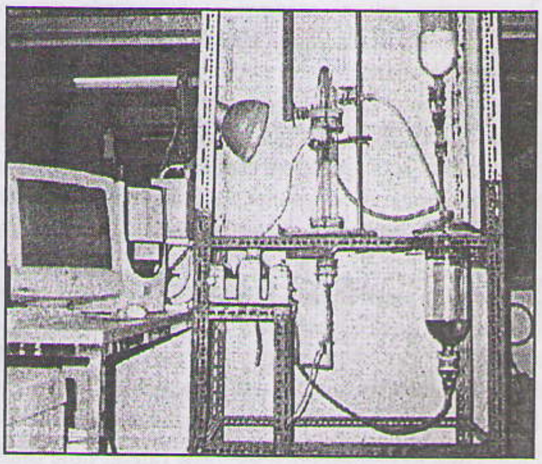

Fig. 1 Photographic view of the equipment

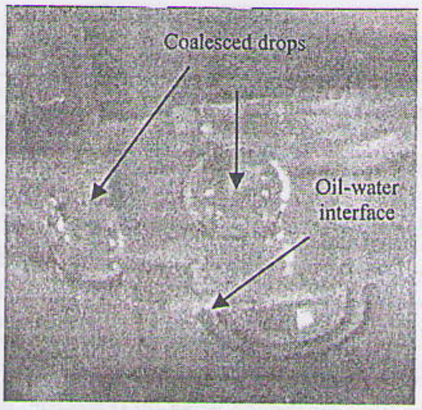

Fig. 2 Photograph of the coalescence column

Photographing equipment

Photography was used to measure time of coalescence while showing drop coalescence at oil-water interface. The camera used was a Surfcam, with USB digital video camera. The lighting was provided by a 100 watt, frosted bulb, reflector type, and flameproof mercury discharge lamp. White reflective plate was put on the side of the column opposite to the camera in order to reflect the light from the lamp, which was, positioned normal to the camera. Thus, lamp light passing through the meniscus near the column wall would be refracted leaving a dark section. To avoid this problem, the light was inclined downward through the liquid-liquid interface. An image viewed at a high incidence angle with the interface would be slightly distorted. This distortion is enhanced during interfacial oscillations that occur during and after impact. To eliminate this problem, the camera was inclined downward slightly to view through the interface from up. The inclination angle was approximately 20°. Since the camera was positioned at an angle, the vertical and horizontal directions were calibrated independently. From the calibration measurements, the actual camera angle was obtained from the following equation:

$$\theta = \cos^{-1}\left(\frac{M_y}{M_x}\right) \qquad (4)$$

Where M_x and M_y were the x-direction and y-direction calibrations in units of pixels/cm, respectively, with an associated uncertainty of 1°. The actual angles were measured directly from a calibration image [17]. The development of the captured image was carried out using computer aided image analysis program of Windows Movie Maker of Microsoft Corporation.

Experimental procedure

At the start of each experiment, the coalescence equipment was cleaned. The glass parts of column were initially degreased with solution of aqueous washed powder, then rinsed with water. The internal sections were cleaned by

Table 1 Physical properties of the system

Material	*Temperature* (°C)	*Density* ($kg\ m^{-3}$)	*Viscosity* ($N\ s\ m^{-2}$)	*Interfacial Tension* ($N\ m^{-1}$)	*API*	*Assay*
Kerosene/Water	30.5	771.82	1.215×10^{-3}	39.45×10^{-3}	49-43	–
	30	995.56	0.84×10^{-3}		–	–
Gas oil/Water	30	810.69	2.125×10^{-3}	43.24×10^{-3}	37	–
	30	995.56	0.84×10^{-3}		–	–
Hexane/Water	30	653	0.345×10^{-3}	53.06×10^{-3}	–	99%
	30	995.56	0.84×10^{-3}		–	–

scrubbing with solution and finally with water. Three sets of experiments were performed using oil (kerosene, gas oil and hexane) as a dispersed phase and water as continuous phase; the physical properties of the liquid-liquid systems employed are summarized in Table 1.

The coalescence column was filled first with distilled water to certain height from the 5-liter QVF glass vessel. Then, the dosing pump was started at certain flow rate and oil from the 5 liter QVF glass vessel was pushed to distributor which included certain nozzle size at bottom of column to create and release the drop to the oil-water interface. The schematic diagram of the whole equipment is shown in Fig. 3.

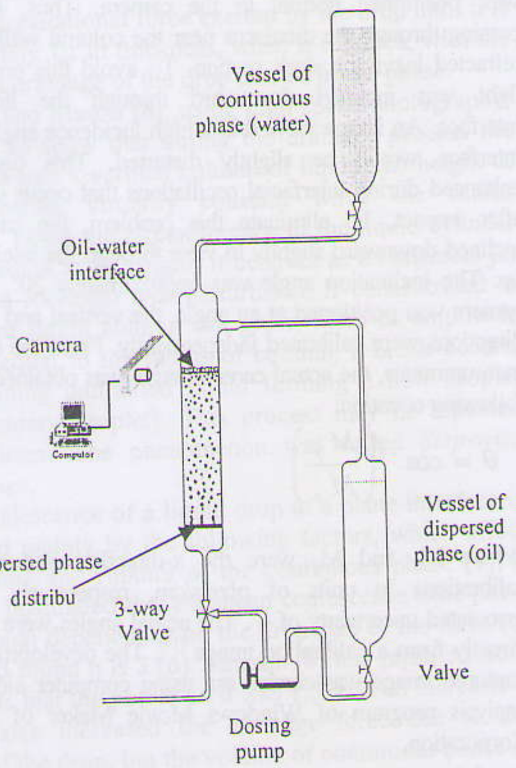

Fig. 3 Schematic diagram of the equipment

About five minutes were given for the equipment to achieve a steady state condition. The camera was synchronized with the lighting to capture the drop coalescence in the illuminated region. The captured images were saved in computer and analyzed to calculate the time of coalescence drop through oil-water interface. The experimental data were obtained in all sets by changing one variable (the height of continuous phase and flow rates of dispersed phase were among the variables changed) while keeping the other constant. Five flow rates were obtained for each height of continuous phase (water height). Hence, distilled water was poured to column to vary the height. At the end value of the height of continuous phase, distributor with other nozzle size was replaced. These steps were carried out at the next set of system and so on, finally all inlet and outlet valves were closed at the same time, and the dosing pump and the camera were switched off.

Results and Discussion

Effect of the dispersed phase flow rate

The experimental results show that increasing flow rate of dispersed phase lead to high increase in the velocity at the nozzles outlet, the dispersed phase liquid issues as a jet and the stream breaks into a large number of small liquid droplets at a short distance from the nozzles. Thus, the increase in flow rate always caused to reduce the drop size whereas the bulk of drops colliding and coalescing with other neighboring drops because of the turbulence in continuous phase. It can be seen clearly that the coalescence time increases with the increase in flow rate of dispersed phase, as it is clarified in Fig. 4-6.

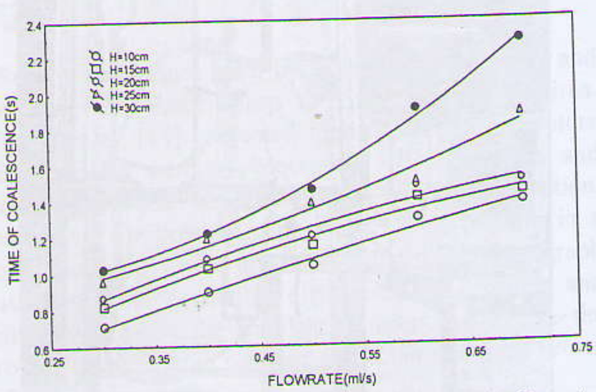

Fig. 4 Time of coalescence vs. flow rate at hole diameter of 3mm for Kerosene/Water System

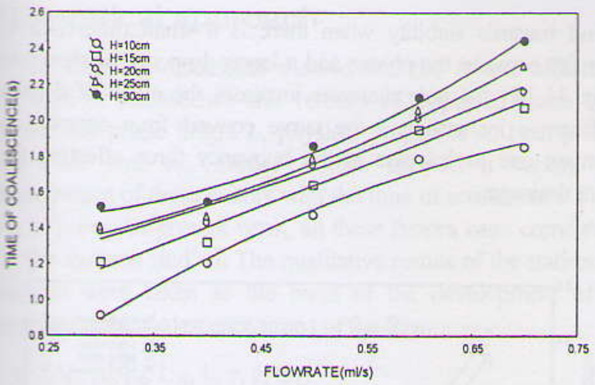

Fig. 5 Time of coalescence vs. flow rate at hole diameter of 3mm for Gas oil/Water System

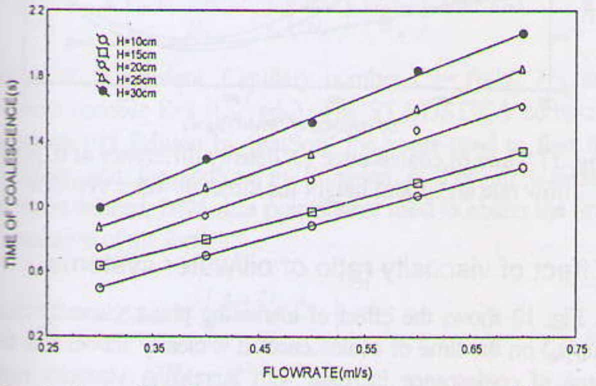

Fig. 6 Time of coalescence vs. flow rate at hole diameter of 3mm for hexane/water system

Effect of continuous phase height

A series of coalescence time determinations were made with the oil/water systems under various conditions to investigate the effect of changing the distance of ascensus of the droplets to the interface. It is observed that time of coalescence increased with increase in the height of continuous phase due to increase in the ascensus of the droplets. The drop released from the hole by uttermost velocity is in column bottom to certain height in the continuous phase. It arrives to terminal velocity at height approximately 20cm. The amplitude of drop size increases when the distance of ascensus increases. This is likely to have an increasing effect on the bouncing of the drop and therefore on the time of coalescence as the distance of ascensus increases. Typical results for three oil/water systems are shown in Fig. 7-9. Variation of coalescence time with height is most likely the result of disturbances caused by impact of the drops with the interface. Such disturbances would increase as the energy possessed by the drops before impact increased that is with increasing length of ascensus, drop size, and the approach velocity of the drop to the interface. It might be thought initially that an increased disturbance would reduce

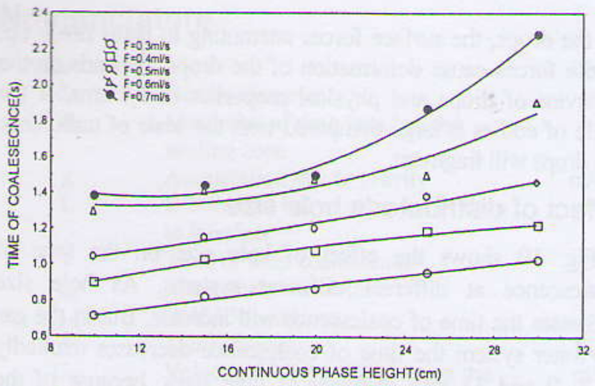

Fig. 7 Time of coalescence vs. continuous phase height at hole diameter of 3mm for kerosene/water system

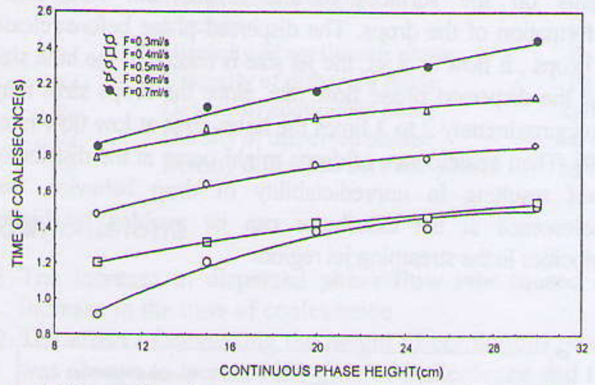

Fig. 8 Time of coalescence vs. continuous phase height at hole diameter 3mm for gas oil/water system

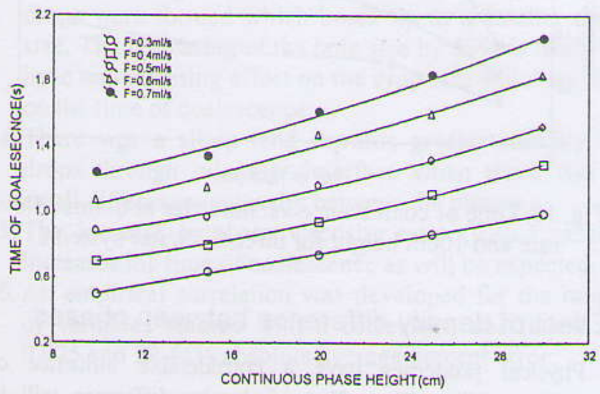

Fig. 9 Time of coalescence vs. continuous phase height at hole diameter 3mm for hexane/water system

the drops stability, but the disturbance would eject the drops from the interface and thereby increase the thickness of the drainage film and consequently time of coalescence. Because of the forces acting on the drops, the dynamic forces due to the turbulent eddies in continuous phase attempting to break up the drops, the surface forces attempting to resist break up. These forces cause deformation of the drops, depends on the behavior of drops and physical properties of systems. If the scale of eddies is large compared with the scale of turbulence the drops will fragment.

Effect of distributer's hole size

Fig. 10 shows the effect of hole size on the time of coalescence at different oil/water systems. As hole size increases the time of coalescence will increase. But in the gas oil/water system the time of coalescence decreases markedly at 8, 9 and 11 mm diameter of hole sizes, because of the fragmentation brought about by the variation in velocity of dispersed phase at nozzles outlet in the turbulent continuous phase, which exert different dynamic pressure at different points on the surfaces of the drops, and also cause deformation of the drops. The dispersed phase before release as drops , it flow as a jet, the jet size is related to the hole size and the dispersed phase flow rate, since the drops sizes may be approximately 2 to 3 times the holes sizes at low flow rates [19]. Then coalescence of drops might occur at the distributor itself resulting in unpredictability of drop behavior, that coalescence at the distributor can be avoided by higher velocities in the streaming jet region.

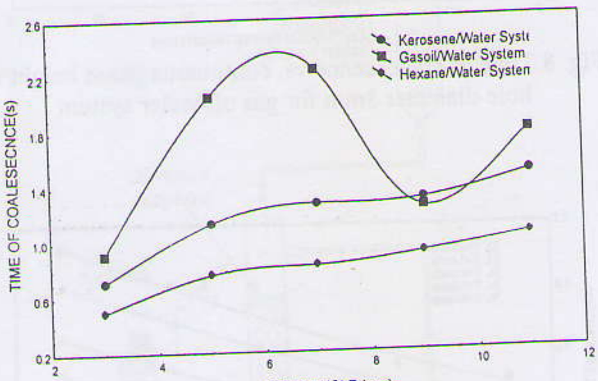

Fig. 10 Time of coalescence vs. hole size at 0.3ml/s flow rate and 10cm height for three oil/water systems

Effect of density difference between phases

Physical properties have a considerable influence on droplets stability. The effect of density difference will be small, because the increased deformation of drop (i.e. flattening of drop) through increase of density difference, will cancel the increase in the drainage force, and some droplets colliding and coalescing with other adjacent droplets, or breaking up either on the oil/water interface, because of generating a wake behind each drop or under the interface, because of the turbulence in continuous phase. This is acting on the results of coalescence time. However, there is a slight trend towards stability when there is a small difference in density between the phases and a larger drop size as shown in Fig. 11. As the hole diameter increases the effect of density difference increase and the curve convert from concave to convex due to increase in the buoyancy force affecting the film drainage.

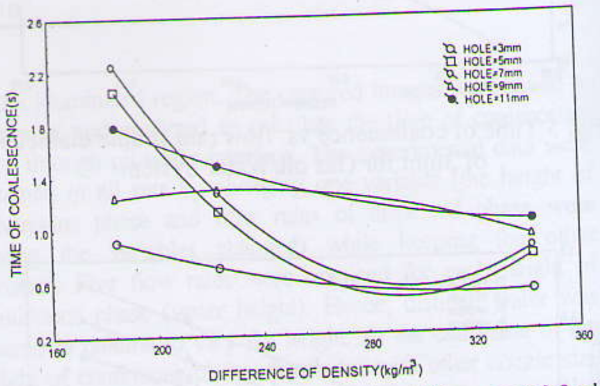

Fig. 11 Time of coalescence vs. density difference at 0.3ml/s flow rate and 10cm height for three oil/water systems

Effect of viscosity ratio of oil/water systems

Fig. 12 shows the effect of increasing phase viscosity rati (μ_d/μ_c) on the time of coalescence. It is clearly shown that th time of coalescence increase with increasing viscosity rati due to increase in the drainage force that assist the rupture c the drop film as will be expected because the resistance t drainage of the film is increased. The time of coalescenc increases, as the hole size increases due to increase in the dro diameter and with higher viscosity, the higher drainage forc obtained. But the reduction in time of coalescence at ho diameter 9 and 11mm and viscosity ratio above 1.8 is due break up of large drop to smaller drop size.

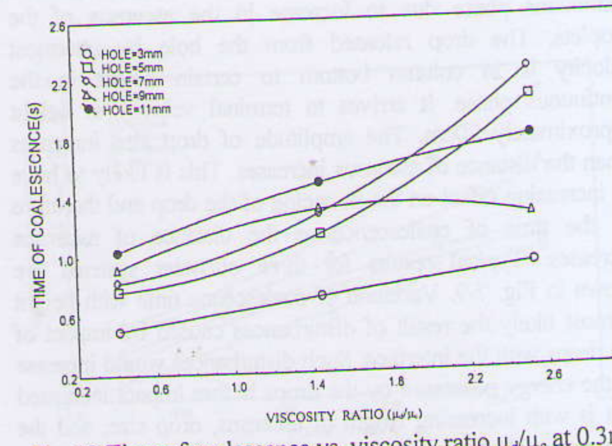

Fig. 12 Time of coalescence vs. viscosity ratio μ_d/μ_c at 0.3 flow rate and 10cm height for three oil/water systems

Theoretical approach

The forgoing discussion showed that the factors affecting the time of coalescence are: velocity of dispersed phase U_N, continuous phase height L, physical properties of both phase and hole size d_N. Earlier studies showed a variety of arrangement of these factors with the time of coalescence [10, 11, 13]. For the present work, all these factors were correlated for the systems studied. The qualitative results of the statistical analysis were taken as the basis of the development of a correlation for coalescence time t of the form:

$$t = f(\Delta\rho, \mu_c, \mu_d, U_N, L, \gamma, g, d_N) \tag{5}$$

Further dimensional analysis showed the correlation in terms of the operating conditions and physical properties is:

$$\frac{U_N\, t}{d_N} = K\left(\frac{\Delta\rho\; U_N\; d_N}{\mu_c}\right)^a \left(\frac{\mu_d}{\mu_c}\right)^b \left(\frac{L}{d_N}\right)^c Ca^d\, Fr^e \tag{6}$$

where K is constant, Capillary number $Ca = (\mu_c U_N / \gamma)$, and Froude number $Fr = (U_N^2 / g d_N)$. The STATISTICA computer program (99 Edition by StatSoft, Inc.) was used to find the constant and the indices in Equation 6. For all oil/water systems studied, 2625 data points were used to obtain the final correlation of the form:

$$\frac{U_N\; t}{d_N} = 9.652\times10^{-3}\left(\frac{\Delta\rho\; U_N\; d_N}{\mu_c}\right)^{1.753} \times\left(\frac{\mu_d}{\mu_c}\right)^{0.79}\left(\frac{L}{d_N}\right)^{0.349} Ca^{-0.032} Fr^{0.021} \tag{7}$$

The scatter of all the experimental data as compared with values calculated from this correlation are almost within ±35%, and with a correlation coefficient of 0.975 and a 22.44% absolute average percent error. These results are shown in Fig. 13.

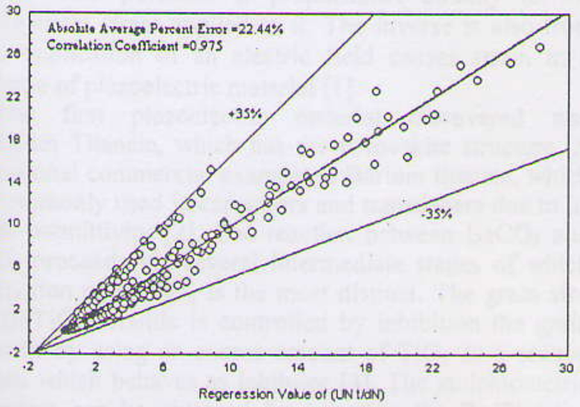

Fig. 13 Time of coalescence correlation for three oil/water systems

Nomenclature

Notation		*Unit*
d_N	Nozzle diameter	m
d_o	Mean size of drop entering the settling zone	m
g	Acceleration due to gravity	m/s^2
L	Distance of fall or ascensus of drop to interface	m
t	Time of drop coalescence	s
t_m	Mean coalescence time	s
$t_{1/2}$	Half-life rest time	s
T	Temperature	°C
U_N	Velocity of dispersed phase at the nozzle	m/s
Greek letters		
γ	Interfacial tension	N/m
ϕ	Inclination angle of lighting	deg.
θ	Actual camera angle	deg.
μ	Viscosity	Pa.s
μ_c	Viscosity of continuous phase	Pa.s
μ_d	Viscosity of dispersed phase	Pa.s
ρ_c	Density of continuous phase	kg/m^3
ρ_d	Density of dispersed phase	kg/m^3
$\Delta\rho$	Density difference between phases	kg/m^3

Conclusions

1. The increase in dispersed phase flow rate caused an increase in the time of coalescence.
2. The effect of increasing the height of continuous phase was always to increase the time of coalescence and the stability of drops. When the drop arrives to the terminal velocity, the amplitude of drop size increases. This is likely to have an increasing effect on the bouncing of the drop.
3. For distributor's hole size greater than 7mm, unstable drops were formed which break up to a smaller drop size. The increasing of the hole size by 51% is likely to have an increasing effect on the drop size and therefore on the time of coalescence.
4. There was a slight tend towards greater stability of drops through oil-water interface when there was a small difference in density between the phases.
5. The increase in phase viscosity ratio (μ_d/μ_c) caused increases the time of coalescence as will be expected.
6. An empirical correlation was developed for the range of variables studied with a correlation coefficient of 0.975 and 22.44% absolute average percent error.

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