Drop Sizes in an Agitated Liquid-Liquid System

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The relationship between drop size and location in an agitated liquid-liquid system was investigated, with a new sampling method in which the dispersion was sampled in a specially designed trap and immediately encapsulated by a polymer film. The liquid-liquid system used was water and a mixture of isooctane and carbon tetrachloride with a density close to that of the water. Dispersed phase holdup was varied from 0.025 to 0.34 volume fraction.

For this system, which has low mutual solubility and high interfacial tension, there is almost no dependence of drop size on location for the mixing geometries studied. This was due to the fact that the coalescence rate is low compared to the circulation time. An increase in impeller speed and drop size decreased the coalescence rate while an increase in holdup increased it.

The mean drop diameter was related to the Weber number and holdup by an equation. By comparison of mean drop diameters obtained using different impellers, it was shown that the criterion of equal power per volume can be used for estimating drop size when going from one mixing geometry to another, not too different, geometry at moderate impeller speeds.

When two mutually insoluble liquids are mixed in an agitated vessel, a dispersion of one phase can be produced in the other continuous phase. Turbulent fluctuations and viscous friction produce forces that tend to breakup the droplets whereas collision between two drops may result in their coalescence into a larger drop. Agitation maintained under constant conditions will result in dynamic equilibrium between the coalescence and breakup processes and appropriate sampling and measurement techniques can be used to determine the size and size distribution of the drops.

A number of investigators have presented equations that relate drop size to mixing parameters and to the physical properties of the liquid system (*1, 2, 11, 16, 19, 22*). Some of the equations relate the mean drop diameter in the vessel to the Weber number and to a hold-up function that represents the secondary effect of the coalescence mechanism. Drop size distribution caused by breakup alone was investigated for mixing vessels for pipes (*4*). Each investigator found a different distribution function suitable to describe his data. It can be said that no general correlation is available at present for the drop size distribution function. Also, the results of studies of single drop breakup (*8, 17, 20*) differ to an extent not fully explained as yet. Divergent results have also been obtained in measurements of coalescence rates in liquid-liquid dispersions due to the different experimental techniques employed (*5, 6, 7*).

Sprow (*19*) and Vanderveen (*21*) studied variation of local mean drop diameter throughout the vessel. They have shown that drop sizes vary in the vessel according to the general model of a circulation path that involves breakup near the impeller followed by continuous coalescence along the circulation path.

DROP SIZE MEASUREMENTS BY ENCAPSULATION

Drop sizes have been determined by a number of techniques including photography, light absorption, and electronic counting (*1, 2, 4, 13, 17, 18, 22*). Most of these methods involved indirect measurements or were limited with respect to location of measurement or operating conditions. The technique that has been developed and used in this work involves sampling of the dispersion followed by immediate encapsulation of the drops with a thin polymer film.

Morgan and Kwolek (*10*) demonstrated that a film of nylon 6-10 is formed at the interface between a solution of sebacyl chloride in carbon tetrachloride and an aqueous solution of hexamethylene diamine. Madden and McCoy (*9*) appear to be the first who applied this interfacial polycondensation technique to encapsulate droplets in an agitated liquid-liquid system. With this technique it is possi-

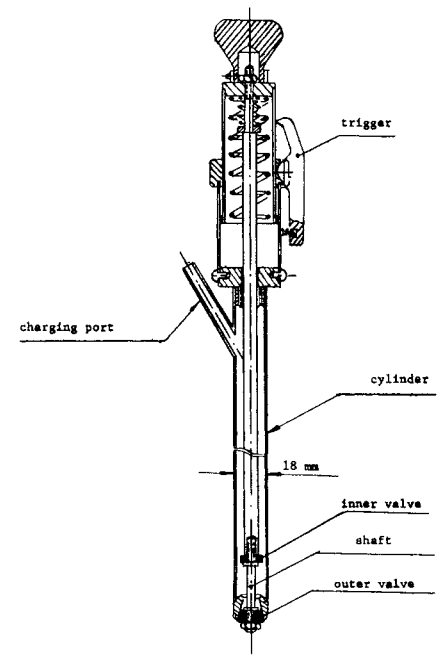

Fig. 1. Trap for sampling and encapsulating drops.

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Table 1. Sampling Point Locations

Sampling point	Radial position, cm.	Height, cm.
1	6	9.6
2	12	9.6
3	12	21
4	3	21
5	1	11
6	6	2
7	12	15

ble to "freeze" the drops in the dynamic conditions they had reached in the system.

In their work, Madden and McCoy encapsulated dispersed-phase carbon tetrachloride droplets in a continuous water phase. Sebacyl chloride was dissolved in the carbon tetrachloride and, following the attainment of steady-state, a small quantity of hexamethylene diamine was added to the mixing vessel and mixing was stopped almost immediately thereafter. Interfacial polycondensation occurred between the two monomers resulting in the encapsulation of the droplets by a thin polymer film. The encapsulated droplets were then sampled and size measurements made by a microscopic technique.

Several drawbacks are inherent in the encapsulation method as described above. Some time is required for the second monomer to disperse throughout the vessel and, during this time, breakup and coalescence are disturbed so that the encapsulated drops may no longer represent the situation that existed in the system prior to the addition of the monomer. Their technique, in addition, does not permit the study of local conditions and only the gross situation throughout the vessel can be analyzed.

LOCAL DROP SIZE MEASUREMENTS BY ENCAPSULATION

In the present work a trapping technique was used which permits reliable sampling at any desired location in the vessel and which overcomes the drawbacks mentioned above. The monomer pair used was piperizine, which is soluble in water, and terephthalic acid chloride, which is soluble in the organic phase. The organic phase in the present study was a mixture of carbon tetrachloride and isooctane.

The trapping device shown in Figure 1 consists essentially of a small cylinder equipped with a spring-actuated double-seal mechanism on an internal shaft. To operate, the spring is depressed so that the cylinder is sealed by the inner valve. A small amount of the continuous phase plus the monomer soluble in it is added to the trap through the charging port. The other monomer had been dissolved in the dispersed phase in the vessel prior to the experimental run. Following attainment of steady state, the trap is placed in the desired sampling position and the trigger is released. The spring mechanism drives the shaft upward so that the vessel contents under the trap enter the cylinder which is then immediately sealed by the outer valve. Interfacial polymerization takes place instantly. Additional drop breakup will not occur because the trapped sample is no longer under the influence of the flow regime and coalescence cannot occur because of the polymer film that now encapsulates the drop.

Preliminary examination of this technique was done in the present study and also by Shiloh et al. (*14, 15*). The drop sizes found using this technique were compared to these obtained by direct photography. The agreement was satisfactory and well within the bounds of the experimental error. The trapping technique was relatively easy to use and met the requirement that it trap and stabilize the individual drops of the dispersed phase as they existed in their normal dynamic situation. The trap, in addition, permits vessel sampling at different locations.

EXPERIMENTAL

Mixing Vessel

The mixing vessel was a pyrex tank, 29 cm. diam., filled with the liquid-liquid system to a height of 29 cm. The vessel was equipped with four equally spaced baffles, 3 cm. in width. A ¼ horsepower variable-speed motor drove the impeller that was located 1/3 of the dispersion height above the bottom of the tank. The impeller was a 6-blade turbine impeller, 10 cm. in diam., blade length 2.5 cm., blade width 2 cm. and disk diam. 7.5 cm. In a few runs a smaller impeller was used. The small impeller was a 4-blade impeller, 9 cm. in diameter, with blade widths of 0.78 cm.

Measurement of Local Drop Sizes

The liquid-liquid system was a mixture of carbon tetrachloride and isooctane that was used as the dispersed phase for most of the runs and distilled water as the second, continuous phase. The density of the organic phase (1.055 g./ml.) was close to that of the water in order to avoid the effect of gravity force on local drop size. Interfacial tension was 41 ± 2 dyne/cm. The dispersed phase was made up with 0.05 wt. % of the monomer soluble in it and the system was agitated for 45 min. to 1 hr. before sampling was begun.

Fig. 2a. Photograph of encapsulated drops.

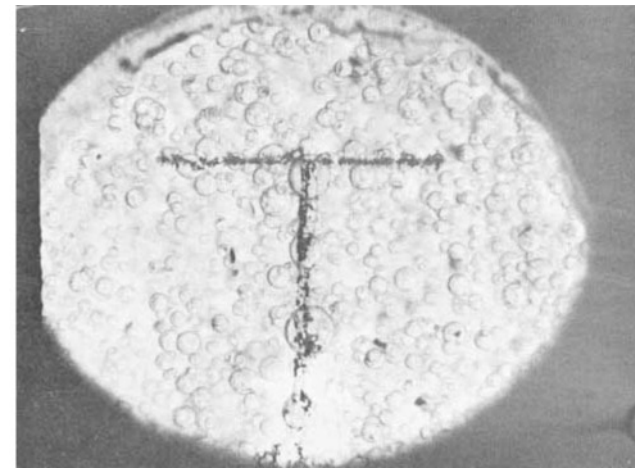

Fig. 2b. Photograph of drops through periscope.

Table 2. Local Mean Diameter

N, rev./min.	Continuous phase	X	d_{32} at sampling location 1	2	3	4	5	6	7
150	Water	0.025	0.325	0.311	0.312	0.340	0.350	0.344	0.317
300	Water	0.025	0.136	0.138	0.140	0.142	0.137	0.123	
500	Water	0.025	0.073	0.082	0.086	0.090	0.077	0.081	
180	Water	0.05	0.290	0.305	0.321	0.390	0.300		0.335
150	Water	0.10	0.440	0.450	0.460		0.470		
300	Water	0.10	0.207	0.240	0.232	0.265	0.229		
500	Water	0.10	0.129	0.130		0.140	0.144	0.134	
200	Water	0.25	0.370	0.314	0.405	0.390	0.411	0.399	0.393
330	Isooctane $+CCl_4$	0.25	0.327	0.334	0.350	0.380		0.346	0.346

In preparation for sampling, some continuous phase material, containing 0.05 wt. % of the monomer soluble in it, was introduced into the sampler cylinder through the charging port. In order to prevent adhesion of encapsulated drops, one to another, in the sample, the continuous phase in the trap contained also a small amount of surface active agent, polyvinyl alcohol when water was the continuous phase, and commercial silicone Rodosil R.7338 when the organic phase was the continuous one. Samples were taken from seven sample locations as described in Table 1. All the points were equally spaced between the baffles (45°).

After trapping, a sample of the encapsulated drops was transferred to a flat glass dish and photographed. Size measurements were made from the enlarged pictures with Carl Zeiss type TGZ 3 Particle Size Analyzer. A typical photograph is shown in Figure 2a.

Coalescence Rate Measurements

Coalescence rates were determined by the same method developed by Doulah and Thornton (5) and Howarth (7) by measuring the initial rate of the mean drop diameter growth after a step-change reduction in impeller speed. This method gives a measure of the coalescence rates at the turbulence level determined by the final impeller speed of drops whose size is determined by the initial impeller speed. It eliminates the drawbacks inherent in other methods which involve mass transfer between phases or require the addition of chemicals as dyes to the dispersion. The models developed by Howarth and Doulah and Thornton related the coalescence rate directly to the initial change in the relative diameter where w, expressed in %/min., can be written as

$$w = 100\left[\frac{1}{d_{32}}\frac{\Delta d_{32}}{\Delta t}\right]_{t=0} \qquad (1)$$

The encapsulation method could not be used for this phase of the investigation because of the need for frequent sampling. The technique used was to photograph the dispersion through a periscope (Figure 3) which consisted of a straight tube closed at the lower end which was fitted with a window and mirror placed at 45°. The periscope was 25-cm. long, 1.3-cm. diam. and the total length between the lens (200 mm. focal distance) and the light sensitive film in the camera was about 68 cm. This arrangement gave about twofold enlargement of the drop image on the photographic film which was not adequate for high agitating speeds or holdup. For this reason experiments were limited to those operating conditions for which good quality photographs were obtained. Figure 2b is a typical photograph as obtained through the periscope.

Table 3. Coalescence Rates

X	N_1, rev./min.	N_2, rev./min.	$(d_{32})_1$, mm.	$(d_{32})_2$, mm.	w, %/min.
		Normal Impeller			
0.025	200	140	0.23	0.37	3.5
0.05	240	180	0.20	0.28	5.1
0.05	180	160	0.28	0.34	4.6
0.10	235	195	0.25	0.32	6.0
0.10	195	160	0.32	0.39	7.5
0.20	235	180	0.35	0.50	8.6
0.34	200	155	0.50	0.67	14.1
		Small Impeller			
0.025	450	385	0.18	0.24	14.5
0.025	385	300	0.24	0.36	8.5
0.025	345	300	0.29	0.35	9.0

Power Measurement

A Cole-Parmer Servodyne was used when power measurements were made. This system gives an electric voltage output which is related only to the moment of the motor and, in addition, gives a constant rotation speed which is independent of the power and can be varied over a wide range.

Experimental Error

The most important of all the factors that may result in measurement errors was the actual measurement of the drop sizes. The static photographs of encapsulated drops were made on samples in glass dishes which contained thousands of drops whereas the area photographed included only a small part of them. Also, the drops tended to congregate in groups randomly so that drop sizes in each group were not necessarily identical

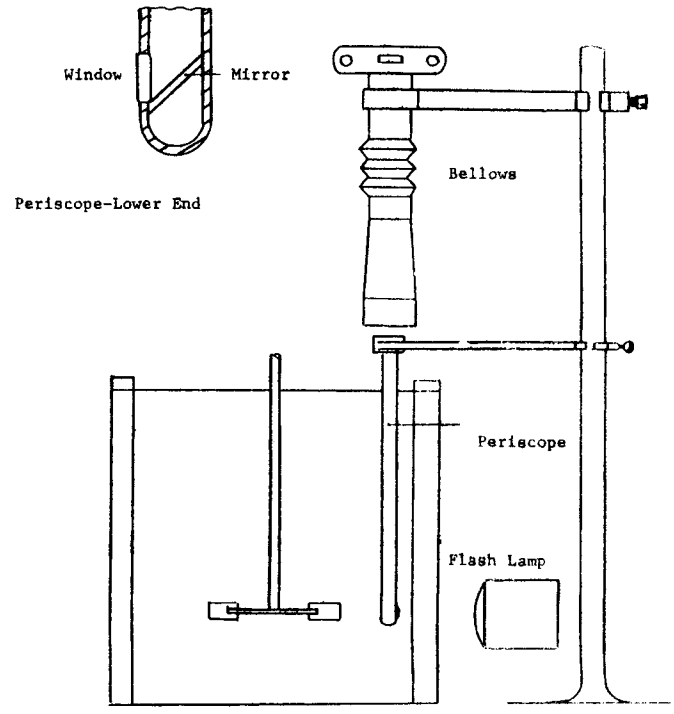

Fig. 3. Mixing system—photos taken through periscope.

to those in all the others. The choice of the area photographed could cause an error because of this randomness. Therefore, two or more photographs were taken from every dish and each picture included at least 300 drops. The drop sizes were determined by averaging all the pictures taken from the same dish. Samples taken from the same location in the vessel at identical conditions had a maximum range of ± 20% in mean drop diameter. In future work it is hoped to eliminate this source of error by developing a technique to photograph the sample in the dish while it is being mixed.

When photos were taken through the periscope, the pictures gave a good representation of the drop population because of the constant flow in front of the periscope window. Two pictures taken one after the other at identical location and conditions gave an agreement of ± 5% in mean drop diameter. On the other hand at high holdups (about 0.1 and greater) or high impeller speeds (200 rev./min. for high holdups and 250 rev./min. for low holdup) the quality of the pictures was not adequate. The small drops, especially, could not be seen clearly. In these cases an agreement of about ± 25% could be obtained between two pictures.

EXPERIMENTAL RESULTS

Local Mean Diameter

The results for the local mean drop diameters, d_{32}, at different locations appear in Table 2. In general, the drops were smaller near the impeller in the jet flowing away from it (point 1, 2) but the variation in local drop size is small.

Coalescence Rate

Coalescence rates were calculated from Equation (1) for experiments made with the normal impeller and with the small impeller. The results are summarized in Table 3. N_1 is impeller speed before the step change was made, $(d_{32})_1$ is the mean drop diameter at steady state before the change, N_2 is the impeller speed after the change and $(d_{32})_2$ is the mean drop diameter at the new steady state.

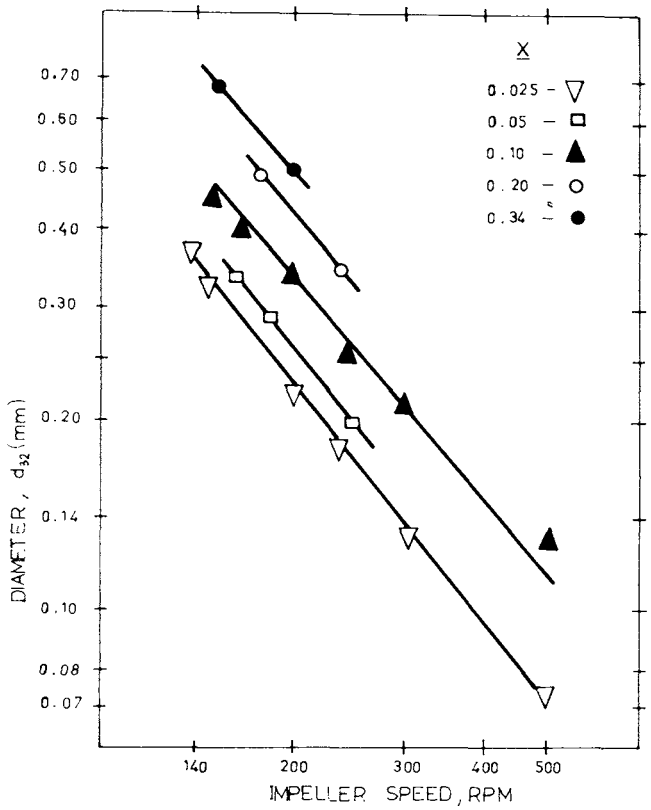

Fig. 4. Drop diameter at steady state.

DISCUSSION

Drop Size at Steady State

The results for mean drop diameter at steady state for different operating conditions are described in Figure 4. The mean drop diameter is related to the impeller diameter, Weber number, and holdup by the equation

$$d_{32}/D = 0.058\ We^{-0.6}\ (1 + 5.4\ X) \qquad (2)$$

This equation, as was shown by Shinnar (*16*), indicates that the kinetic breakup mechanism is the main factor in determining drop sizes. The secondary effect of the coalescence mechanism is represented by the holdup function

$$f(X) = 1 + 5.4\ X \qquad (3)$$

The linear form of the holdup function fits the data well for $X \leq 0.2$. For higher holdup the dependence of d_{32} on X is smaller. The same behavior was reported by Brown and Pitt (*1*) for kerosene in water and $X > 0.3$. Although another form of the holdup function might fit the data better, including the data for higher holdup, this linear form was preferred because fewer measurements were made for high holdup and, in addition, the accuracy of the measurements was lower. The linear form also permitted the present results to be compared to those obtained by other investigators. Vermeulen et al. (*22*), Calderbank (*2*), and Brown and Pitt (*1*) found the following values, respectively, for the coefficient of X: 2.5, 9, and 3.14.

Brown and Pitt (*1*) suggested, in contrast to the common notion, that the holdup function does not represent a coalescence effect but a damping of the turbulence caused by the presence of the dispersed phase. They based this argument on their findings that showed that the same holdup function could describe the behavior of a highly coalescing liquid-liquid pair as well as that of a low coalescing pair. They also suggested that the high value of the coefficient of X as reported by Calderbank (*2*) was the result of measuring drop sizes not at the impeller tip but remote from it. Nevertheless, it seems that there are cases where drop sizes at the impeller tip are influenced by the drop sizes of the dispersion flowing into the impeller zone. This drop population does coalesce to an extent which depends largely on the holdup, as was shown separately in the coalescence rate experiments.

Coalescence Rates

Coalescence rates as calculated from Equation (1) appear in Table 3. The least square technique was used to correlate the data for the normal impeller and the following expression resulted:

$$w \propto N^{-1.8}\ X^{0.8}\ d_{32}^{-0.9} \qquad (4)$$

Although this equation is only preliminary because the small number of data points available did not permit an accurate evaluation of the exponents, the equation does show the trends and interrelationships among the parameters. The coalescence rate decreased markedly with an increase in impeller speeds as was found by Doulah and Thornton (*5*) for the isooctane water system they studied. The inverse dependence on the drop diameter has been shown to correspond to the case where coalescence takes place in a kinetic flow regime when cohesive forces between the drops are weak (*14*).

Circulation rates were calculated from the impeller pumping capacity (*12, 13*) and were found to be high as compared to the coalescence rates. Circulation times of the order of 5 to 10 sec. were calculated as compared to coalescence rates of the order of 5 to 10 %/min. As a result a significant variation of local drop sizes cannot be ex-

pected for this system. The effect of increased circulation times on the coalescence rate was studied by changing to a smaller impeller which had a larger circulation time than the normal impeller. The result in Table 3 for 140 rev./min. with normal impeller should be compared to those for 300 rev./min. with small impeller. For these two cases, the average drop diameters were the same and power consumptions were equal but the pumping rate of the small impeller was one-half that of the normal impeller, that is, the circulation time was doubled. The coalescence rate also increased by a factor of 2.4. The coalescence rates with the small impeller were still too small, however, to effect a significant variation in local drop sizes.

For liquid-liquid systems having low interfacial tension the coalescence rates were found (*5*) to be an order of magnitude higher. Also the variation of local drop sizes as reported by Vanderveen (*21*) was more pronounced for systems with low interfacial tension. Those findings agree well with the results reported here and also with those observed by Brown and Pitt (*1*).

Taking into account the circulation time effect it was concluded that, for the same liquid pairs, local drop size variation may increase or decrease depending on the geometry of the mixing system. Further research in this direction is of great importance as it can provide more helpful and necessary information for design and scale-up purposes.

Power as a Scale-up Criterion

The comparison of input powers for the two impellers when producing the same mean drop diameter is summarized in Table 4. N_1, P_1 are, respectively, impeller speed and power for the normal impeller and N_2, P_2 are for the small impeller. From these results it can be concluded that the criterion of equal power per unit volume can be applied quite well for estimating mean drop diameter when going from one mixing geometry to another, not too different geometry. The criterion is not so clear, however, for the higher impeller speed.

It is known that the power as a possible scale-up criterion is only one among others. The criterion to be chosen depends on operating conditions and on the relative effects of the breakup and coalescence mechanisms. If coalescence is of lesser importance more appropriate criterion could be the impeller speed alone, because, as the impeller speed is increased, both circulation time and coalescence decrease. One can then expect a shift toward a condition where the impeller speed is the preferred criterion. Such a case was indeed reported by Brown and Pitt (*1*) for a mixing system similar to that of the present study but at higher speeds and shorter circulation times which resulted not only from the impeller speed but also from the greater blade widths of the impellers used in their study.

Encapsulation Method

Most of the data in Figure 4 was obtained using the encapsulation method. The agreement between Equation (2) and the results obtained by other investigators (*1, 2, 22*) is another indication of the reliability of this method. It should be pointed out that the encapsulation method is especially advantageous and suitable for size distribution measurements. In the present study, however, more attention was given to the variation in local mean drop diameter and its relation to the coalescence rate. Also the experimental error, which can be reduced considerably as mentioned before, did not justify a thorough analysis of size distribution at this stage. In addition, the error in measuring the larger drops is much smaller than for the smaller drops which have only a small effect on the mean diameter, d_{32}, as used in this work.

Table 4. Power Comparison for Two Impellers

d_{32}, mm.	N_1, rev./min.	N_2, rev./min.	P_1, (in.) (lb.)(sec.)$^{-1}$	P_2, (in.) (lb.)(sec.)$^{-1}$
0.18	240	450	12.5	9.5
0.24	180	385	5.2	5.4
0.29	160	345	3.6	3.6
0.36	140	300	2.4	2.3

Although the encapsulation method is easy to use and the trapping technique permitted information to be obtained on drop sizes at any desired location in the vessel, one disadvantage was that some liquid-liquid pairs examined did not give encapsulation of the drops. The pairs examined were water as one phase and organic liquids with relatively high solubility in water and low interfacial tension as the second phase. The organic liquids examined were alcohols from n-butanol to n-octanol, methyl isobutyl ketone and cyclohexanone. More attention to this problem may result in finding the liquid-liquid pairs, not necessarily with water as one of the phases, with low interfacial tension which can be satisfactorily encapsulated. Such pairs would enable an extension of this study to pairs with low interfacial tension.

SUMMARY

A new sampling technique was used in the present study. The encapsulating method with the trapping technique proved to be reliable and easy to use. It was found that some liquid pairs are not eligible for encapsulation with the monomers used and this factor calls for more research in the direction of selection of eligible encapsulating monomers. The liquid-liquid system in the present study, with results typical for its high surface tension, exhibited small variation in local drop sizes due to low coalescence rates relative to the circulation time of the dispersion.

The mean drop diameter was correlated by Equation (2) which is in good agreement with results obtained by other investigators who used different experimental techniques.

ACKNOWLEDGMENT

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NOTATION

D = impeller diameter
d_{32} = Sauter mean drop diameter
N = impeller speed
P = input power
t = time
X = holdup
$f(X)$ = holdup function, Equation (3)
w = coalescence rate expressed as relative rate of change of diameter
We = $\rho N^2 D^3/\sigma$, Weber number

Greek Letters

ρ = density
σ = interfacial tension

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Distributed-Parameter Dynamics by Correlation Analysis

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The experimental dynamic response of a distributed-parameter, simultaneous heat and mass transfer system was investigated using correlation analysis. The system was a wetted-wall column operating as a nonadiabatic humidifier with both liquid and gas phases in turbulent flow.

The experimental frequency response results were compared to the values predicted by an existing mathematical model. The investigation also included a study of the effect of employing forcing functions having nonideal power spectra. Analog-simulated, first-order systems were tested to verify the computational procedure and to support the conclusions for the humidification system.

System dynamic identifications, acceptable for most engineering purposes, were obtained from forcing signals having power spectra which differed significantly from ideality. For the reduction technique employed, a time-domain record of approximately 2,500 pairs of input-output data points was found to suffice for a satisfactory analysis. The correlation technique yielded reliable results over approximately the same range of frequencies as reported for previous pulse test studies based on a comparable forcing procedure. However, the results showed an upper frequency limit below that previously achieved by direct frequency forcing of the same system.

The dynamic characteristics of chemical process systems are often obtained from differential equations derived from appropriate mass, energy, and momentum balances. However, if the process is too complex to permit mathematical modeling, or if verification of a model is desired, experimental testing techniques may be employed. Common experimental testing methods are direct frequency forcing, pulse testing, transient step response analysis, and correlation analysis.

The correlation technique employs a very complicated forcing function which may be considered to be the realization of random noise. In this sense, the term random is commonly used loosely to imply an irregular variation with time that cannot be predicted in advance.

The most attractive feature of correlation analysis is its validity for open-loop systems in the presence of corrupting noise which is uncorrelated with the system forcing function. Thus the technique is not limited, as are other experimental methods, by the necessity of maintaining all other process variables at steady values during the test. Another advantage is the possibility of finding a suitable disturbance already present in the input variable such that the process need not be artificially upset. Limitations of correlation analysis include the necessity of processing large amounts of data and the danger of steady state drifts yielding nonstationary functions which are unsuitable for system identification.

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