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MASS TRANSFER IN A CONTINUOUS-FLOW MIXING VESSEL

by

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MASS TRANSFER IN A CONTINUOUS-FLOW MIXING VESSEL

by

Virgil G. Trice, Jr.

ABSTRACT

Individual continuous-phase film coefficients have been measured on an area basis for the transfer of mass between partially miscible liquids in a continuous-flow mixing vessel. For the systems studied (isobutanol dispersed in water and nitrobenzene dispersed in ethylene glycol) the mass transfer coefficient, in agreement with the penetration theory, was found to vary as the square root of the molecular diffusivity. The applicability of a previously suggested relationship was demonstrated by the correlation of the experimental data in terms of the Sherwood, Schmidt, and Reynolds numbers. The relationship obtained was in fundamental agreement with correlations developed by investigators using other types of liquid-liquid contacting apparatus.

SUMMARY

In this investigation, the transfer of mass between liquid phases was studied in a continuous-flow mixing vessel. Experimental data were obtained for the partially miscible systems isobutanol in water and nitrobenzene in ethylene glycol. These data were used to test the applicability of contemporary film theory and to determine the validity of suggested methods of correlation.

Satisfactory experimental techniques were developed to determine individual film coefficients for the continuous phase on an area basis. The interfacial areas of the disperse systems were determined from light transmittance measurements. The concentration differences were measured by sampling the extract phase of the disperse system in the mixing vessel. Preferential sampling of the extract phase was accomplished by means of porous glass filters.

For the systems studied the interfacial area produced by flow through a mixing vessel was found to be a function of vessel diameter, dispersed phase flow rate, and the volume fraction of the dispersed phase:

$$(N_{Re})_d = f \left\{ (x_v)^{1/2} (N_{We})^{2/3} / N_{Re} \right\} \quad (6)$$

The experimental data were in agreement with the suggestion that film coefficients for mass transfer in mixing vessels may be correlated by a relationship involving the Sherwood, Schmidt, and Reynolds numbers. Based on the experimental results of the present investigation the following equation is recommended for the continuous phase film coefficient:

$$(N_{Sh})_d / (N_{Sc})^{1/2} = 0.03 (N_{Re})^{0.28} \quad (12)$$

The relationship recommended is in agreement with correlations developed by investigators using other types of liquid-liquid contacting apparatus. In addition, it is similar to, but not in exact agreement with, correlations developed for the dissolution of solid pellets in an agitated vessel.

The results of this investigation lend further experimental confirmation to the applicability of the penetration theory developed by Higbie and Danckwerts in that the mass transfer coefficients obtained were found to be a function of the square root of the molecular diffusivity.

I. INTRODUCTION

Despite the increasing emphasis on liquid-liquid extraction, very little work applicable to the design of industrial contactors has been reported in terms of fundamental mechanisms.⁽¹⁷⁾ Current design methods provide essentially rough approximations based in many cases on empirical relationships limited to specific systems and on comparison with existing units of similar design.

There can be little question about the need for a more fundamental approach to the study of mass transfer in dispersed systems of immiscible liquids. This need was emphasized some time ago at Argonne National Laboratory by an abortive attempt to make a choice between solvents for a mixer-settler extraction system based on the physical properties of the system. It was apparent from that experience that progress in this field was seriously impeded by the lack of satisfactory experimental methods for measuring interfacial area and for preferential phase sampling. Accordingly there was set up a three-part program:

1. to develop a rapid and accurate method for estimation of the interfacial area of liquid-liquid dispersions,
2. to correlate the interfacial area formed in agitated* vessels with the physical and geometrical parameters of the system, and

*It is necessary to distinguish between mixing effected by means of mechanical agitation and by means of turbulence produced only by the kinetic energy of the influents. Therefore, in this report, the term "agitated" is restricted to the former case wherein mixing is effected by the action of an impeller.

3. to use the information obtained in (1) and (2) to contribute to the understanding of the fundamentals of mass transfer between immiscible liquids in mixing vessels.

Portions of the results of the first two parts of this program have been published.^(12,13,18,20) This report deals with the third part. It is hoped that the results of this work will stimulate other investigators to offer fundamental contributions in this area of unit operations.

II. GENERAL CONSIDERATIONS

In dealing with liquid-liquid extraction the rate of mass transfer is usually expressed in terms of a mass transfer coefficient K (see Sect. VII for the notation employed):

$$N = KA (c^* - c) \quad (1)$$

The absence of satisfactory experimental techniques for measuring the interfacial area, A , and concentration difference, $c^* - c$, has prevented the determination of mass transfer coefficients for liquid-liquid systems in continuous-flow mixing vessels. They have been measured, however, by means of single-drop studies⁽⁵⁾ and in certain types of equipment where methods were available for measuring the interfacial area. The latter include: spray towers,⁽⁷⁾ wetted-wall columns,⁽¹⁾ and gently agitated chambers⁽⁶⁾ and horizontal pipes⁽¹¹⁾ with relatively undisturbed interfaces.

Rushton and various co-workers are responsible for much of our present knowledge of the fundamentals of fluid mixing. In a survey, Rushton⁽¹⁵⁾ briefly considered the transfer of mass between immiscible liquids and suggested the following correlation for mass transfer data:

$$(N_{Sh}) (N_{Sc})^{-s} = f(N_{Re}) \quad (2)$$

Although this equation has not been experimentally verified for liquid-liquid systems in mixing vessels, there is considerable evidence which indicates that it represents the proper relationship between the pertinent fluid motion and mass transfer parameters. For example, satisfactory correlations of this type were obtained by Hixson and Baum⁽⁹⁾ for liquid-solid systems in an agitated vessel and by Heertjes, et al.⁽⁷⁾ and Ruby and Elgin⁽¹⁴⁾ for liquid-liquid systems in a spray tower. Mention should also be made of the analogy with heat transfer since extensive use has been made of relationships of this type.

Mass transfer presents a more complex picture in liquid-liquid than in liquid-solid systems. Both continuous and discontinuous-phase film resistances must be considered. Furthermore, fluid motion affects not only the mass transfer coefficient, but also the mean drop size of the

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dispersion. Murphy and co-workers,⁽¹¹⁾ recognizing the complex nature of the problem, introduced additional dimensionless groups including the viscosity ratio and a group involving length, interfacial tension, viscosity, and diffusivity. Experimental evidence was presented which showed the additional groups to be significant parameters for mass transfer in a horizontal pipe.

In order to conform to the limitations of time and manpower, it was necessary to concentrate on a small portion of the overall problem. At the same time, it was desired to provide a program to test the applicability of the film theory and to compare results with those obtained by other investigators in studies of liquid-solid systems. Accordingly, the experimental program was confined to the measurement and correlation of the continuous-phase film coefficients for the partially miscible systems, isobutanol in water and nitrobenzene in ethylene glycol. This particular pair of systems was chosen because of their widely differing molecular diffusivities.

Individual film coefficients may be obtained by a technique originally described by Colburn and Welsh.⁽²⁾ Briefly stated, this technique is based on the assumption that for a binary system of limited mutual solubility there exists only one film resistance for each of the species transferring. For example, in any oil-water system, the oil would have to penetrate a water film only. The validity of this technique is well established and the results of the present investigation can be used to deduce further confirmation.

For the special case of the mixing vessel where partially miscible phases are contacted and the organic phase is both dispersed and introduced at saturation:

$$k_c = \frac{N}{A(c^* - c)} \quad (3)$$

In this case mass transfer is unidirectional, with the resistance to transfer in the aqueous film. The mass transfer coefficient k_c is an individual film coefficient in units of cm/sec. The interfacial area (A) and the concentration difference ($c^* - c$) are those values which actually exist in the mixing vessel during steady-state operation. Since the contents of the chamber are thoroughly mixed to give a uniform dispersion, the compositions and flow-rates of the chamber contents and the effluent stream are identical.

III. EXPERIMENTAL

Experimental data were obtained in four- and six-inch, baffled mixing vessels. The influents were pumped with centrifugal pumps from 160-gal reservoirs to the mixing vessel. Adequate influent rates were used to disperse the organic phase uniformly without the aid of mechanical agitation. The two-phase effluent from the mixing vessel was discharged to a weigh tank which also served as a holdup tank for phase separation. The raffinate

effluent was recycled to the influent reservoir whereas the extract effluent was discarded. Where necessary a raffinate effluent clean-up step was employed to remove any surface active contamination.

A schematic flow diagram is shown in Figure 1. A pressure of 3 to 5 psi was maintained in the reservoirs to provide a positive suction head and to facilitate pump priming. The influent flowrates were controlled by throttling valves and measured with calibrated manometers. A 20-gal, electrically heated still was provided for solvent clean-up.

FIGURE 1
SCHEMATIC FLOW DIAGRAM

A. Mixing Vessels

A detailed sketch of the 4-inch vessel is shown in Figure 2. The vessel formed a right cylinder with a brass wall and top flange. Closure of the cylinder base by means of a disk of 1/2-inch plate glass with a matching brass flange permitted observation of the mixing pattern and entry of the light beam for interfacial area determination. Effluents were discharged through two ports. These ports were located near the top and bottom of the chamber to minimize short-circuiting effects. Four vertical baffles, each approximately one-tenth the diameter of the chamber, were used.

FIGURE 2
SKETCH OF FOUR-INCH MIXING VESSEL

Influents, introduced at the center line, impinged on a horizontal distributor. Openings were provided in the top flange for a thermometer and a light probe (used for interfacial area measurement). Satisfactory liquid seals for glass rods were obtained with modified Swagelok fittings. Fittings were bored out to the required diameter and the stainless steel insert replaced with Teflon.

The measured volumes of the four- and six-inch units, including the discharge ports, were 950 and 3050 cu cm, respectively. The two units, based on design suggestions of Rushton,⁽¹⁵⁾ were dimensionally similar, but differed in construction in that glass cylinder walls were used in the six-inch vessel.

B. Sampling

The concentration difference in equation (1) is the difference between the saturation composition and the actual composition of the extract phase in the mixing vessel. In the procedure usually employed for sampling emulsions, the phases of the dispersion in an aliquot are first separated by settling and subsequently a specimen of the desired phase is withdrawn for analysis. Realizing that some mass is transferred during

the settling period, some investigators have sought to obtain the true composition in the mixing vessel by making corrections to the compositions determined by this method. At best this procedure provides a poor method of sampling and was useless in the present work because of the rapid rates of mass transfer for the systems studied. Therefore, in order to determine accurately the composition of the extract phase in the mixing vessel, it was necessary to develop a procedure for sampling preferentially the desired phase of an emulsion.

A technique for the separation of phases of liquid-liquid emulsions by means of porous membranes was described by Jordan.⁽¹⁰⁾ The ability of a membrane to discriminate between phases was attributed to pore size, wetting characteristics, interfacial tension between the liquid pair, and pressure drop. In the present work the continuous phase (extract) was sampled successfully by means of "D" porosity Ace glass filters. For continuous phase sampling the filter was cleaned with acid-dichromate solution and thoroughly washed with water to insure preferential wetting by the aqueous* phase. Samplers intended for organic phase sampling were coated with a thin layer of silicone, using Dri-Film. Attempts to sample preferentially the dispersed organic phase were not successful. This is not to say that it could not be accomplished by this method. However, it may be inferred from comments in Jordan's paper that it would be difficult if not impossible to sample a dispersed phase preferentially. Fortunately, the data desired for the systems studied required the sampling of only the continuous phase.

A number of preliminary experiments were made to test Ace glass filters as a means of aqueous phase sampling using a one-to-one dispersion of water-saturated isobutanol in water. Tests were made using pressure differentials across the filters of 5 to 30 psi and filters of A, B, C, and D porosity. The A, B, and C filters were unsatisfactory, although the C filter did produce samples about 75 per cent aqueous at lower pressures, indicating some phase discrimination. The phase selectivity of the D filters varied over a wide range, depending on the porosity of the particular specimen. Some D filters were very porous and passed liquid as readily as many of the C filters, while one D filter in the test batch did not pass any liquid at 30 psi. As anticipated, the degree of phase discrimination varied inversely with the porosity. About 20 per cent of the D filters tested demonstrated nearly complete phase discrimination at 25 psi, producing 100 per cent aqueous samples with an occasional drop of organic appearing in every third or fourth sample.

*The term "aqueous" refers to the more polar liquid which was employed both as the continuous phase and the extract. In the system nitrobenzene-ethylene glycol, "aqueous" refers to the ethylene glycol phase.

C. Physical Property Measurements and Analytical Techniques

The physical properties of the systems studied are given in Table 1. Densities were determined pycnometrically, viscosities by means of Ostwald viscosimeters and Brookfield rotational viscosimeters, interfacial tensions by means of the drop-weight method, refractive index at the sodium D line with an Abbe refractometer, and diffusivities were calculated by means of the Wilke correlation.⁽²¹⁾

Analytical techniques were developed for determining the isobutanol content of water refractometrically and the nitrobenzene content of ethylene glycol colorimetrically.

Refractive index is plotted as a function of composition for solutions of isobutanol in water in Figure 8 and for nitrobenzene in ethylene glycol in Figure 9 (see Appendix). Although refractive index measurement left much to be desired in terms of accuracy and precision as an analytical method for isobutanol, its use was dictated by the lack of a suitable alternative.

For determining nitrobenzene in ethylene glycol, refractive index measurements were totally unacceptable and a much more precise colorimetric technique was developed. A few experimental measurements showed that at 260 millimicrons a linear relationship existed at low concentrations between nitrobenzene composition and optical density. A Beckman DU Spectrophotometer with 0.1-cm cells was used. The greatest accuracy of the spectrophotometer was in the optical density range from 0.163 to 0.825, corresponding to a maximum nitrobenzene molarity of 9.74×10^{-4} with an average molarity to optical density ratio of 1.18×10^{-3} . The necessary 1000-fold dilution of the run samples, which had molarities in the order of 0.8, was accomplished with distilled 95 per cent ethyl alcohol.

D. Measurement of Interfacial Area

The interfacial areas of the liquid-liquid dispersions were determined from light transmittance measurements. The equipment and techniques are described in detail in a paper by Trice and Rodger.⁽¹⁹⁾ Briefly, the equipment consisted of a microscope-lamp light source with a red filter and an 18-mm aperture, a platinum-coated glass rod light probe, and a phototube (RCA 1P41)* together with the necessary electronic equipment for the detection and amplification of the phototube output. Correlations are available for transparent organic liquids dispersed in water which give interfacial area as a function of optical density for 50, 25, and 10 volume per cent dispersed phase. These correlations, Figure 11 (see Appendix), were used to determine the interfacial area of dispersions of isobutanol in water.

*The red filter, Corning 2404, together with the 1P41 phototube provided an effective light spectrum in the near infrared region.

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Table 1

PHYSICAL PROPERTIES OF SYSTEMS STUDIED^a

System	Isobutanol in Water	Nitrobenzene in Ethylene Glycol
Density of drop phase, g/cu cm	0.8339	1.2144
Density of field phase, g/cu cm	0.9859	1.1160
Viscosity of drop phase, g/cm sec	0.0293	0.0185
Viscosity of field phase, g/cm sec	0.0120	0.143
Interfacial tension, dynes/cm	2.2	6.4
Diffusivity, ^b sq cm/sec	8.28×10^{-6}	6.36×10^{-7}
Refractive index of drop phase, $n_D^{(25)}$	1.3884	1.5489
Refractive index of field phase, $n_D^{(25)}$	1.3406	1.4391

Source of Materials

Isobutanol	- Commercial grade, DeNert and Dougherty, Inc., Chicago, Illinois
Ethylene Glycol	- Low conductivity grade, C. P. Hall Company, Chicago, Illinois
Nitrobenzene	- Commercial grade, E. I. DuPont De Nemours and Company, Gibbstown, New Jersey
Water	- Local supply tap water

^aPhysical properties are for mutually saturated liquids at 25C.^bDiffusivities were calculated by means of the Wilke correlation.⁽²¹⁾

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Although nitrobenzene has a strong yellow color it is transparent in the near infrared region. However, since the nitrobenzene was dispersed in ethylene glycol and not water, the previously mentioned light transmittance-interfacial area correlation does not apply. As shown in Figure 10 (see Appendix), there is a difference in the light absorbancy of ethylene glycol and water in the spectrum of interest. Therefore, a series of runs were made in a six-inch agitated vessel to obtain data necessary for an interfacial area-light transmittance correlation for 50, 25, and 10 per cent dispersions of nitrobenzene in ethylene glycol. Satisfactory correlations, developed for 25 and 10 per cent dispersions, are presented in Figures 12 and 13 (see Appendix). Unfortunately, photographs of the 50 per cent dispersions revealed a phase inversion with small nitrobenzene drops appearing in the larger ethylene glycol drops and the interfacial area could not be defined. It is interesting to note that both correlations are functions of AL , the product of interfacial area and optical path length, rather than of $AL^{0.65}$ as is the case with the correlations for transparent liquids dispersed in water.

E. Procedure

The operating sequence may be summarized as follows:

1. Introduction of the continuous (extract) phase to wet the sampler and to provide a base line corresponding to 100 per cent light transmission:
2. Introduction of the dispersed (raffinate) phase and adjustment of flowrates, and
3. Sampling and measurement of light transmittance during the steady state portion of the run.

Equations (1) and (3) are defined in terms of the effluent flow rates. As previously mentioned the effluent flow rates and compositions are the same as those in the mixing vessel. They were determined indirectly from the influent flow rates or from time-weight measurements of the effluents.

Runs were made at individual phase flow rates up to 3.0 gpm. Organic to aqueous flow ratios were varied from 1/9 to 1/1. Steady-state conditions, as indicated by light transmittance measurements, were realized almost instantaneously. During the steady-state portion of the run, the aqueous phase was sampled continuously at a rate of about one cu cm per minute, and light transmittance measurements were taken at approximately two-minute intervals. The operating time under steady state conditions varied from 10 to 15 min depending on the flow rates employed.

IV. CORRELATION OF EXPERIMENTAL DATA

The purpose of this work was two-fold: first, to develop experimental techniques for measuring the individual mass transfer coefficient for the continuous phase as defined by the equation

$$N = k_c A (c^* - c) \quad , \quad (3)$$

and second, to determine whether the mass transfer coefficient may be related to the pertinent fluid motion and physical property parameters by the equation

$$(N_{Sh})/(N_{Sc})^S = f(N_{Re}) \quad . \quad (2)$$

The experimental data are summarized in Tables 2 and 3.

A. Interfacial Area in Continuous-Flow Mixing Vessels

When two immiscible liquids are pumped through a mixing vessel at flow rates greater than some threshold value a disperse system is obtained. In order to apply equation (3) the interfacial area of this disperse system must be known. In most cases, if the organic to aqueous flow ratio is less than unity, the organic phase will be dispersed. In the great majority of cases, however, the interfacial area for water-in-oil systems cannot be defined because of the presence of small oil drops in the water drops.⁽¹⁸⁾ For that reason only organic to aqueous flow ratios of unity or less were used in this work, and the organic phase was always dispersed.

The interfacial area of dispersions of immiscible liquids can be determined photographically.⁽¹⁹⁾ Light transmittance-interfacial area correlations are available for transparent liquids dispersed in water.⁽²⁰⁾ For transparent liquids dispersed in liquids other than water a light transmittance correlation can be developed for that particular system provided that the light absorbency of the field phase liquid is low. The latter procedure was used in this work for the nitrobenzene-ethylene glycol runs.

Some work has been reported for non-flow conditions on interfacial area in agitated vessels as a function of fluid motion.⁽¹³⁾ A correlation was developed which gave the interfacial area as a function of a number of physical and geometric variables. This correlation cannot be used as a means of predicting the interfacial area in a continuous-flow vessel; however, it does emphasize the complex nature of the problem. Glycol-nitrobenzene mixing data, obtained in a non-flow agitated chamber in connection with the light transmission-interfacial area runs, did not fit the correlation given for non-flow conditions. However, this was not a valid test because the glycol to nitrobenzene viscosity ratio far exceeded the experimental limits of the correlation.

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Table 2

SUMMARY OF EXPERIMENTAL DATA FOR ISOBUTANOL-IN-WATER RUNS

Run	Flow Rate (gpm)		Volume Fraction Dispersed Phase, x_v	Interfacial Area, a (sq cm/cu cm)	Run Temp (C)	Composition Effluent Water, c (g Isobutanol/L)	Concentration Difference, $c^* - c$ (g Isobutanol/L)	Mass Transfer Coefficient, k_c (cm/sec)
	Field Phase	Dispersed Phase						
6-in. Vessel								
1A	1.0 ^b	1.0	0.50	41.5	26.5	d	<1.7	-
2A	1.5	1.5	0.50	90.5	27.0	d	<1.7	-
3A	2.0	2.0	0.50	104.5	26.0	d	<1.7	-
4A	2.4	2.4	0.50	110.0	26.0	d	<1.7	-
5A	2.8	2.8	0.50	125.5	27.0	d	<1.7	-
6A	3.0	3.0	0.50	128.5	27.0	d	<1.7	-
7A ^c	3.2	3.2	0.50	154.5	27.0	d	<1.7	-
8A ^c	3.0	3.0	0.50	145.0	24.0	d	<1.7	-
9A ^c	3.0	3.0	0.50	148.5	26.0	d	<1.7	-
10A ^c	3.4	3.4	0.50	154.5	27.0	d	<1.7	-
11A ^c	2.8	2.8	0.50	139.5	25.0	d	<1.7	-
12A ^c	3.0	3.0	0.50	150.0	26.5	d	<1.7	-
13A	3.0	3.0	0.50	132.0	26.0	d	<1.7	-
14A	3.0	3.0	0.50	126.5	26.5	d	<1.7	-
4-in. Vessel								
15A ^a	1.5	1.5	0.50	93.0	26.5	70.0	8.4	0.893 x 10 ⁻²
16A	1.5	1.5	0.50	97.1	27.0	d	<1.7	-
17A	2.0	2.0	0.50	95.0	26.5	d	<1.7	-
18A	2.5	2.5	0.50	131.5	26.5	d	<1.7	-
19A	3.0	3.0	0.50	148.5	26.5	d	<1.7	-
20A ^a	3.0	1.0	0.25	83.0	21.0	76.9	5.9	3.11 x 10 ⁻²
21A ^b	1.0	1.0	0.50	-	24.0	-	5.0	-
22A ^b	1.0	1.0	0.50	-	27.0	-	4.2	-
23A	3.0	0.33	0.10	40.5	27.7	75.0	2.9	10.5 x 10 ⁻²
24A	3.0	0.33	0.10	39.5	23.0	75.0	5.9	6.41 x 10 ⁻²
25A	3.0	0.33	0.10	37.0	26.0	69.7	9.2	4.08 x 10 ⁻²
26A	3.0	1.0	0.25	76.0	26.7	76.7	1.7	11.8 x 10 ⁻²
27A	2.5	0.29	0.104	35.0	27.9	72.4	5.0	6.87 x 10 ⁻²
28A	2.5	0.625	0.20	55.0	27.9	74.1	2.5	8.86 x 10 ⁻²
29A	2.5	1.07	0.333	74.0	27.5	d	<1.7	-
30A	2.5	1.66	0.25	92.5	27.5	d	<1.7	-

a Sampler failure suspected

b Flow rates too low to provide complete emulsification

c Surface-active contamination present

d Within limits of analytical precision, extract effluent was isobutanol-saturated.

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Table 3

SUMMARY OF EXPERIMENTAL DATA FOR NITROBENZENE IN ETHYLENE GLYCOL RUNS
(4-IN. VESSEL)

Run	Effluent Flow Rate (gpm)		Volume Fraction Dispersed Phase, x_v	Interfacial Area, a (sq cm/cu cm)	Run Temp (C)	Composition Effluent Glycol, c (g Nitrobenzene/L)	Concentration Differences, $c^* - c$ (g Nitrobenzene/L)	Mass Transfer Coefficient, k_c (cm/sec)
	Field Phase	Dispersed Phase						
1B	1.96	0.41	0.173	28.0	28.0	46.0	46.0	4.65×10^{-3}
2B	2.04	0.59	0.224	35.8	28.0	55.7	36.3	5.80×10^{-3}
3B	2.04	0.97	0.322	47.7	27.5	70.7	21.3	9.42×10^{-3}
4B	2.08	1.40	0.402	63.8	27.0	81.0	11.0	15.94×10^{-3}
5B	2.03	1.65	0.448	71.0	27.0	78.7	13.3	11.23×10^{-3}
6B	2.05	0.85	0.293	55.4	28.0	75.5	16.5	11.24×10^{-3}
7B	Sampler Failure							
8B	2.07	0.65	0.239	45.4	28.0	59.5	32.5	5.54×10^{-3}
9B	1.98	0.47	0.195	38.5	26.0	50.9	41.1	4.23×10^{-3}
10B	1.96	0.44	0.183	30.8	28.0	45.7	46.3	4.17×10^{-3}
11B	2.04	0.88	0.301	52.0	28.0	65.6	26.4	6.47×10^{-3}
12B	2.06	1.43	0.410	81.0	27.5	81.9	10.1	13.69×10^{-3}
13B	2.31	0.82	0.262	57.5	26.0	66.1	25.9	6.81×10^{-3}
14B	1.75	0.66	0.274	46.2	27.8	60.5	31.5	4.83×10^{-3}
15B	1.45	0.55	0.275	2.93	28.2	53.9	38.1	4.65×10^{-3}
16B ^a	2.03	1.45	0.417	81.8	27.0	72.9	19.1	6.29×10^{-3}
17B	2.00	0.58	0.225	41.0	28.0	59.6	32.4	5.96×10^{-3}
18B	1.35	0.53	0.282	29.8	28.5	49.1	42.9	3.44×10^{-3}

^aSampler failure suspected

The problem of relating the interfacial area produced to fluid motion is simpler for the continuous-flow chamber than for the non-flow agitated chamber. Since only two systems were studied it was not possible to establish a general correlation, but it was possible to determine the effect of vessel diameter, flowrate through the chamber, and volume fraction of the dispersed phase.

For the systems studied, the interfacial area produced in a continuous-flow mixing vessel appeared to be independent of the continuous phase flowrate. To provide a typical example, the data obtained for the isobutanol-water system in a four-inch vessel are plotted in Figure 14 (see Appendix). As may be seen, a smooth curve was obtained by plotting the interfacial area, in sq cm per cu cm of total mixed phases, against the dispersed phase flowrate, in gpm. The continuous phase flow rate does not appear to have any significant effect although it was varied by a factor of two from 1.5 to 3.0 gpm. The effect of vessel diameter is apparent from the data of Table 2. For a given flow rate the interfacial area roughly varies inversely to the $1/4$ power of the vessel diameter.

From the preceding discussion, it is apparent that the fundamental variables are interfacial area (a), dispersed phase flow rate (q_d), the vessel diameter (T), and possibly the volume fraction of the dispersed phase (x_v). It is convenient to define several other variables which are functions of the fundamental variables:

$$V_d = q_d / \frac{\pi T^2}{4} \quad (4a)$$

$$d = 6 x_v / a = \frac{\sum n_i d_i^3}{\sum n_i d_i^2}, \quad x_v = \frac{V_d}{V_d + V_c} \quad (4b)$$

$$N_{Ar} = \frac{\mu_f a}{V_d \rho_f} \quad (4c)$$

$$N_{Re} = \frac{T V_d \rho_f}{\mu_f} \quad (4d)$$

$$N_{We} = \frac{T V_d^2 \rho_f}{\sigma} \quad (4e)$$

$$N_{Dr} = \frac{q_d \rho_f}{\mu_f d} \quad (4f)$$

$$(N_{Re})_d = \frac{d V_d \rho_f}{\mu_f} \quad (4g)$$

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These groups were used to show the interfacial area or the mean drop diameter as a function of the fluid motion parameters. Two equations are presented. In the order of decreasing average deviations they are

$$N_{Ar} = f [N_{Re} / (N_{We})^{2/3}] \quad (5)$$

$$(N_{Re})_d = f [N_{Re} / (N_{We})^{2/3} x_v^{1/2}] \quad (6)$$

Equation (5) follows logically from the experimental evidence that neither the continuous-phase flow rate nor the volume fraction appear to have any effect on the interfacial area. Figure 15 (see Appendix) shows a plot of equation (5). The average deviation for the nitrobenzene-ethylene glycol data is about 10 per cent with a maximum deviation of about 20 per cent. The isobutanol-water data give a slightly better correlation.

An improved correlation may be obtained by introducing the volume fraction of the dispersed phase as an independent variable. As may be seen from equation (4b) this procedure implies that the continuous-phase flow rate has a significant effect which could not be detected from the raw data. It might be mentioned that the definition of x_v [equation (4b)] in terms of flowrates has been a controversial point with some investigators. However, the validity of this relationship was demonstrated by Flynn and Treybal⁽⁴⁾ and was experimentally verified in the present work.

In Figure 3, a Reynolds number based on the mean surface-volume drop diameter is shown as a function of V_D , T , and x_v as given by equation (6). Equation (6) gives the best correlation with an average deviation in drop diameter of about five per cent and a maximum deviation less than 20 per cent.

In considering these deviations, it should be pointed out that the interfacial areas used in these correlations were based on light transmittance measurements and that the general correlation of light transmittance with interfacial area has an average deviation of 5 per cent. In addition, experimental light transmittance data for the nitrobenzene-glycol system were not available for dispersed phase volume fractions greater than 0.25. It was necessary, therefore, to extrapolate these data to volume fractions as great as 0.45.* Runs 15B, 18B and 1A, Tables 2 and 3, were not considered in calculating the average deviations, because the flow rates employed were too low to provide uniform dispersions.

*Interpolation and extrapolation of light transmittance data may best be accomplished by plotting the log of the volume fraction vs the interfacial area for a given value of I_0/I .

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FIGURE 3
REYNOLDS NUMBER BASED ON MEAN DROP DIAMETER AS A FUNCTION
OF FLUID MOTION PARAMETERS

It is reasonable to expect that an analogy should exist between the interfacial area produced by fluid motion in a continuous-flow mixing vessel and in an agitated chamber of similar design. Unfortunately, the relationship developed for agitated vessels is based on a different method of correlation. However, as previously mentioned, the continuous-flow vessel presents a simpler problem and, in fact, the correlation developed for the present work suggests what may be a better method for correlating agitated chamber data.

The dimensionless groups $(N_{Re})_d$, N_{Re} , and N_{We} may be defined for the agitated chamber if the logical assumption is made that the superficial velocity (V_d) may be replaced by the product of impeller speed and diameter, nD .

$$(N_{Re})_d = \frac{dnD \rho_f}{\mu_f} \quad (7a)$$

$$N_{We} = \frac{T (nD)^2 \rho_f}{\sigma} \quad (7b)$$

$$N_{Re} = \frac{T nD \rho_f}{\mu_f} \quad (7c)$$

Data for any specified liquid pair should correlate in the following form:

$$(N_{Re})_d = f [N_{Re} / (N_{We})^b x_v^h] \quad (8)$$

Sufficient agitated chamber data are available to form a generalized correlation which could be used to aid in the development of a generalized correlation for the interfacial area formed in a continuous-flow vessel.

B. Mass Transfer Coefficient as a Function of Fluid Motion and the Physical Properties of the Liquid Pair

As previously mentioned, it has been suggested by Rushton⁽¹⁵⁾ that a generalized correlation may be obtainable for the mass transfer coefficient in the following form:

$$N_{Sh} / (N_{Sc})^s = f(N_{Re}) \quad (2)$$

The original film theory as postulated by Whitman indicated an exponent of zero for the Schmidt group. However, the more recently developed Higbie-Danckwerts penetration theory^(8,3) is supported by the experimental work of a number of investigators. This theory indicates that the mass transfer coefficient should be proportional to the square root of the molecular diffusivity. The present work gives additional support to the value of 1/2 for the exponent s .

The experimental conditions of the present work were designed to facilitate an analogy between liquid-liquid and liquid-solid mass transfer. In liquid-solid mass transfer, mass is transferred from a solid pellet through a field-phase film to the bulk continuous phase. In the present work, however, mass was transferred from an aqueous-saturated organic drop through a field-phase film to the bulk continuous phase.

Hixson and Baum⁽⁹⁾ successfully correlated mass transfer coefficients for liquid-solid systems in an agitated vessel by means of equation (2). They defined the dimensionless groups as follows:

$$\frac{k_c T}{D_v} / \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} = f \left(\frac{n T^2 \rho_f}{\mu_f} \right) = f(N_{Re}) \quad (9)$$

Recent mixing theory is not in agreement with the definition given in equation (9) for the Reynolds number. Fundamental principles require that the impeller diameter appear in the mixing Reynolds number, and the current practice is to use as the characteristic length the impeller diameter D rather than the tank diameter T . Exception may be taken with current practice because analogy with the case of the continuous-flow mixing vessel, equation (7c), suggests the definition of the Reynolds number in an agitated vessel to be

$$N_{Re} = \frac{n D T \rho_f}{\mu_f}$$

This point does not represent a valid argument with the correlation of Hixson and Baum because they obtained their experimental data at a constant ratio of impeller to tank diameter. It is reasonably certain, however, that this correlation would not hold for different size impellers in a given agitated vessel.

Another questionable point is the omission of the particle diameter as a pertinent variable. In single drop and in spray-tower studies the particle diameter has been used as a characteristic length.⁽⁷⁾ This would seem to be a reasonable approach because, in dealing with film phenomena, both particle size and velocity have been found to be significant variables.

For comparison with the correlation of Hixson and Baum, equation (2) is defined for mass transfer in a continuous-flow mixing vessel as follows:

$$\frac{k_c T}{D_v} \left/ \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} \right. = f \left(\frac{T V_d \rho_f}{\mu_f} \right) \quad (10)$$

The mean drop diameter may be satisfactorily introduced in two ways:

$$\frac{k_c T}{D_v} \left/ \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} \right. = f \left(\frac{T V_d \rho_f}{\mu_f} \right) \left(\frac{T}{d} \right) = f \left(\frac{q_d \rho_f}{\mu_f d} \right) \quad (11)$$

and

$$\frac{k_c d}{D_v} \left/ \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} \right. = f \left(\frac{T V_d \rho_f}{\mu_f} \right) \quad (12)$$

Equations 10, 11, and 12 are plotted in Figures 4, 5, and 6, respectively.

FIGURE 4
CORRELATION OF MASS TRANSFER COEFFICIENT BY METHOD
OF HIXON AND BAUM FOR LIQUID-SOLID SYSTEMS

FIGURE 5
CORRELATION OF MASS TRANSFER COEFFICIENT
WITH DROP DIAMETER FUNCTION

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FIGURE 6
CORRELATION OF MASS TRANSFER COEFFICIENT WITH
REYNOLDS NUMBER OF DISPERSED PHASE (RECOMMENDED)

V. DISCUSSION

The correlations presented are based on experimental data obtained in a total of 48 runs. Thirty runs were devoted to the study of the system isobutanol in water. The six-inch mixing vessel was used for 15 of these runs. The four-inch chamber was used for the remainder of the isobutanol-water runs and for the 18 nitrobenzene-ethylene glycol runs.

A. Interfacial Area

The continuous-flow mixing vessel presents the least complicated case for a fundamental study of mass transfer in commercial-type equipment. It has the advantage that the concentration difference does not vary with time and that the compositions of the effluents are the same as the compositions of the phases in the mixing vessel. The one factor largely responsible for preventing the effective use of this tool has been the problem of measuring the interfacial area of disperse systems of immiscible liquids. In the present work this problem was handled by the use of a light transmittance-interfacial area correlation or by photographing the dispersion. Neither method is entirely satisfactory. Light transmittance can be used only for transparent liquids, and the determination of interfacial area from photographs is a time-consuming laborious art.

A general correlation of interfacial area as a function of fluid motion in a continuous-flow mixing vessel is very definitely needed. It was not possible to develop a general correlation from the results of the present work because only two liquid systems were studied. There is the possibility, however, that by comparison with the data for interfacial area in agitated vessels a correlation could be obtained by analogy.

Some conclusions are suggested by the data with respect to the effect of certain variables on the interfacial area. The important independent variables are the flow rate of the dispersed phase, the vessel diameter, and the volume fraction of the dispersed phase. Equation (6) leads to a reasonable relationship between the variables and is recommended as the best method of correlating these data. It may be noted from Figure 3 that different slopes were obtained for each system, as was the case with agitated chamber data.⁽¹²⁾ The effect of the variables on the interfacial area (a) may be summarized as follows:

Variable	Effect on Interfacial Area (a)	
	Isobutanol-water	Nitrobenzene-ethylene glycol
Vessel Diameter, T	$a \sim T^{-0.25}$	$a \sim T^{-0.37}$
Dispersed phase flowrate, q_d	$a \sim q_d^{0.42}$	$a \sim q_d^{0.46}$
Volume fraction dispersed phase, x_v	$a \sim x_v^{0.13}$	$a \sim x_v^{0.19}$

The indicated difference in the effect of vessel diameter for the two systems studied might be anticipated from a comparison of the field-phase viscosities. The viscosity of the ethylene glycol phase was about 10 times that of the water phase. Previous work with agitated vessels⁽¹³⁾ suggested that the velocity of the drop at the wall of the vessel has considerable effect on drop breakup. This means, of course, that the greater the tank diameter the larger the mean drop diameter and, therefore, the lower the interfacial area for a given dispersed phase flowrate. It also follows that the greater the field-phase viscosity the greater the deceleration of the drop and the lower the interfacial area for a given dispersed phase flow rate. The latter conclusion is confirmed by the experimental data. For example, in the four-inch vessel at a dispersed phase flow rate of one gpm, about 20 per cent more area was produced in the isobutanol-water runs than in the nitrobenzene-ethylene glycol runs.

Isobutanol-water runs 7A through 12A were not used in the correlation because of the inadvertent contamination of the isobutanol phase with some unknown surface-active material. The effect of this surface-active contamination, shown in Figure 7, was to increase the interfacial area

FIGURE 7
EFFECT OF SURFACE-ACTIVE CONTAMINATION
ON INTERFACIAL AREA
(Isobutanol-Water System in 6-in Chamber)

about 15 per cent for a given flow rate. Very little is known about the role of surface-active contamination; however, the ratio of dispersion settling times to the $1/6$ power was found to be the most sensitive measure of the effect of contamination in agitated systems.⁽¹²⁾ Rough measurements were made of settling time for Runs 8A and 14A. The dispersed phase flow rate was 3.0 gpm in both cases. The settling time factor for 8A, using contaminated solvent, and 14A, using purified solvent, was $(1.08/0.76)^{1/6}$ giving a predicted increase in area due to surface-active contamination of 6 per cent.

B. Mass Transfer

Satisfactory data for the calculation of mass transfer coefficients were obtained from 6 of the 30 isobutanol-water runs and 16 of the 18 nitrobenzene-ethylene glycol runs. These data were used to test equation (2) as a method of correlating mass transfer coefficients for liquid-liquid systems in a continuous-flow mixing vessel.

In the first series of runs, Runs 1A through 15A, the isobutanol-water system was studied in the glass-walled six-inch vessel. Influent flow rates, which would provide a measurable concentration difference for the calculation of mass transfer coefficients, were estimated from spray-tower

and wetted-wall column data. These calculations erroneously indicated that satisfactory data could be obtained in the six-inch vessel at all influent flow rates above 1.0 gpm. Accordingly, the mixing vessel and associated facilities were designed for a maximum of 3.0 gpm and a glass-walled mixing vessel was used to facilitate observation of the mixing pattern.

The rate of mass transfer, however, was found to be much greater than anticipated. Based on the correlations developed in the present work, the estimate of satisfactory influent rates for the isobutanol-water system was grossly in error. Within the limits of analytical precision, the extract effluents were solute-saturated. With an operating pressure in the glass mixing vessel of 22 psi at 3.0 gpm it was not considered safe to increase the flow rates further. Although the rate of transfer of nitrobenzene to ethylene-glycol was much lower, it was not used in the six-inch vessel because of the extreme toxicity of nitrobenzene. Moreover, since the viscosity of glycol is 10 times that of water, higher operating pressures would have been required.

For the reasons mentioned, the data obtained in the six-inch vessel were satisfactory for use in an interfacial area correlation but not for the calculation of mass transfer coefficients. In recognition of the practical limit imposed by pump capacity and solvent inventory on the flow rates that could be employed, a smaller four-inch metal-walled mixing vessel was constructed. All experimental data used in the mass transfer correlations were obtained in the four-inch vessel.

In considering the mass transfer correlations presented it should be remembered that only the individual film coefficient for the continuous phase was considered. When, for example, a third component or solute is present which transfers from one phase to the other, two film resistances must be considered. The present work, therefore, cannot be used to predict the overall mass transfer coefficient except in the special case where the continuous-phase film offers the principal resistance. Moreover, the present work does not apply if the transferring species undergoes a chemical reaction at the interface. Further comments on this subject are made in the Section V.C.

As may be seen from the experimental data, Tables 2 and 3, knowledge of the interfacial area is necessary in order to explain the phenomenon of mass transfer in a continuous-flow mixing vessel. For example, assuming a constant organic to aqueous flow ratio, an increase in flow rate would be expected to result in a decrease in the per cent saturation of the extract effluent. However, this is not the case because the area increases approximately as the square root of the dispersed phase flow rate and the mass transfer coefficient increases directly with flow rate. It is important to remember that the preceding statement does not apply to an agitated vessel but to the case being considered where turbulence is supplied only by the kinetic energy of the influents.

The isobutanol-water data provide an excellent example of the effect of flow rate. At high dispersed-phase flow rates, within the limits of analytical and sampling precision, the extract effluent was solute-saturated. By decreasing the dispersed-phase flow rate the per cent saturation of the extract effluent was decreased to the extent that the concentration difference could be measured.

The liquid systems studied provide a rigorous test of equation (2) as a correlating tool in that the Schmidt numbers differ by a factor of more than 100. Figure 4 was obtained by defining the dimensionless parameters in a manner analogous to that used by Hixson and Baum for liquid-solid systems. In Figures 5 and 6, which lead to approximately identical correlations, the drop diameter was included as a significant variable.

The correlations show surprisingly little scatter of the data, considering the difficult nature of the sampling problem. As previously pointed out, it was necessary to sample the extract phase of the mixing chamber contents in the presence of drops of the raffinate phase. It must be assumed that the porous glass filters, which were used as samplers, did not effect total phase discrimination even though there was no visual evidence to the contrary. A small amount of sample contamination would have a lesser effect on the nitrobenzene-ethylene glycol data, because the extract phase effluents were always less than 90 per cent saturated. On the other hand, the extract effluent from the isobutanol-water runs was always greater than 85 per cent saturated.

Another factor which contributed to the scatter of the data was the precision of the analytical techniques. The colorimetric method for nitrobenzene gave results reproducible to within ± 2.0 per cent. Therefore, the maximum possible error in the concentration difference, attributable to the analytical technique, would be ± 20 per cent for a 90 per cent saturated glycol effluent. For the isobutanol-water system, the possible error in isobutanol concentration was ± 0.84 gm per liter. This means the possible error in the concentration difference, ascribable to analytical error, ranged from 7 to 50 per cent. The upper limit was arbitrary and simply means that data were not included in the mass transfer correlations unless the indicated concentration difference was greater than 1.7 gm per liter.

The correlations definitely show that, for the systems studied, the individual film coefficient for the transfer of mass through the continuous-phase film may be defined by the relationship given in equation (2). Furthermore, the analogy with similar correlations based on data from wetted-wall columns and spray towers confirms the use of the Schmidt number to the $1/2$ power and suggests the generality of the relationship.

There remains, however, the task of selecting the proper definitions for the Sherwood, Schmidt, and Reynolds groups. The problem in defining the dimensionless parameters is the proper choice of the characteristic

length. Hixson and Baum⁽⁹⁾ working with liquid-solid systems employed the vessel diameter. Investigators studying mass transfer from single liquid drops to a field liquid and mass and heat transfer from single solid spheres to a field fluid have used the particle diameter.⁽¹⁶⁾ For the conditions of the present work it seems reasonable to assume that both the drop and the vessel diameter should appear in a generalized correlation.

The correlations based on equations 11 and 12, which include both the drop and vessel diameters, lead to virtually identical relationships. Also, the use of the drop diameter as the characteristic length in both the Reynolds and Schmidt groups leads to essentially the same relationship as the Hixson and Baum method. The two basic correlations reduce to the following relationships:

$$\text{Hixson and Baum method, } \frac{k_c T}{D_v} \left/ \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} \right. = 3.3 \left(\frac{T V_d \rho_f}{\mu_f} \right)^{1.1}$$

$$k_c \sim \frac{q_d^{1.1}}{T^{2.1}} \quad (10a)$$

and equation (11) to

$$\frac{k_c T}{D_v} \left/ \left(\frac{\mu_f}{\rho_f D_v} \right)^{1/2} \right. = 5.53 \times 10^{-2} \left(\frac{q_d \rho_f}{\mu_f d} \right)^{0.91}$$

$$k_c \sim \frac{q_d^{0.91}}{T d^{0.91}}$$

Incorporating the equivalent for the mean drop diameter as defined for the nitrobenzene in ethylene glycol system in Figure (3),

$$k_c \sim \frac{q_d^{1.3}}{T^{1.34} x_v^{0.74}} \quad (11a)$$

By means of equations (10a) and (11a) it may be shown that the Hixson and Baum method does not provide the best correlation of the experimental data. However, mention should be made of the fact that both correlations predict concentration differences too small to measure experimentally ($\Delta c < 0.84$ g/L) for the series of isobutanol-in-water runs in the six-inch vessel. In that respect both are in agreement with the experimental data. The significant point is that equation (11a) includes the dispersed-phase volume fraction, thereby indicating that the continuous-phase flow rate has an effect on the mass transfer coefficient.

Equations (10a) and (11a) are compared in Figure 16 (see Appendix) by means of the nitrobenzene in ethylene glycol data. It is apparent that equation (11a) provides the better correlation. Neglecting Runs 6B and 4B, deviations from equation (11a) do not exceed 20 per cent, whereas for equation (10a) the deviations are as great as 40 per cent.

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Equations 11 and 12 provide a more logical relationship between the pertinent variables. Equation (12) is in the form suggested by Rushton. It is recommended as the preferred method for the correlation of individual continuous-phase film coefficients for the transfer of mass between immiscible liquid phases in a continuous-flow mixing vessel. The restriction remains, however, that the transferring species does not react chemically at the interface.

C. Suggestions for Future Work

It would be desirable to test further equation (12) as a method of correlating continuous flow mass transfer data. There is a need for data obtained in other sizes of mixing vessels, an extension of the range of mean drop size, and the acquisition of data for other liquid pairs.

The generality of the recommended correlation cannot be firmly established without data for other sizes of mixing vessels. However, reduction of vessel diameter to sizes less than four inches introduces the specter of wall effect. While contemplating an increase in vessel diameter, technical and economic requirements associated with large solvent inventories and pump capacities must be considered. In connection with the latter, it should be remembered that there are, for a given diameter vessel, minimum flow rates below which a uniform dispersion cannot be obtained. Although methods are not available for estimating the minimum flow rates, it might be expected that, in general, the larger the vessel the greater the minimum flow rate.

Previous work in agitated chambers with a large variety of liquid pairs showed that the range of mean drop sizes which may be produced by conventional methods of agitation was limited to about a factor of five. The present work confirms this observation. As may be seen from Table 4 (see Appendix) despite the range of flow rates and flow ratios employed, the maximum variation in mean drop sizes for either system was only a factor of two. From previous experience it may be predicted that for partially miscible systems in a continuous-flow mixing chamber the mean drop size will rarely exceed 600 or be less than 100 microns. Therefore, since it is not likely that the range of mean drop sizes can be extended, other methods must be employed to test the mass transfer correlation.

For the two systems studied the molecular diffusivities and continuous-phase viscosities differed by more than a factor of 10 and the Schmidt numbers by more than a factor of 100. These figures represent very nearly the extremes that can be obtained by means of partially miscible liquids. Few, if any, systems, with the exception of nitrobenzene-ethylene glycol, provide Schmidt numbers as high as 200,000. For most of the partially miscible systems, isobutanol-water being a typical example,

the Schmidt number is of the order of 1,000 to 10,000. These systems, characterized by high rates of mass transfer, can provide useful data provided sufficient emphasis is placed on the accuracy of the sampling and analytical techniques. Since the physical properties are functions of temperature, it would be advantageous to obtain data over as wide a temperature range as possible.

A useful extension of the present work would be the study of individual film coefficients for the dispersed phase. One possible approach would be the study of unidirectional mass transfer from a saturated field phase to a partially saturated dispersed phase. In this case the dispersed phase would be the extract phase. Preferably, the organic phase should be dispersed in order to avoid a water-in-oil emulsion, because satisfactory procedures are not currently available for defining the interfacial area of these systems.(18)

In the present work some consideration was given to the measurement of film coefficients of the dispersed phase. One of the reasons why this effort was not pursued was the lack of a satisfactory method for preferentially sampling the dispersed phase. Attempts to sample by means of silicone-coated porous glass filters were unsuccessful. The use of porous Teflon appeared to offer some promise; however, this approach was not thoroughly investigated.

A second method for measuring dispersed-phase film coefficients involves the use of a third component or solute. The continuous-phase film coefficient could be calculated by means of equation (12). Assuming the additivity of film resistances, the dispersed-phase film coefficient could then be determined by difference. Film coefficients determined by the method described could be compared with those obtained using partially miscible liquids. This would provide a good test of the well-established concept of the additivity of film resistance. The use of a third component would also permit an evaluation of the effect of the direction of mass transfer on the mass transfer coefficient.

The suggestions for future work are briefly summarized as follows:

- A. Obtain additional data to test adequately the correlation recommended for the continuous-phase film coefficient.
 1. Investigate the effect of vessel diameter.
 2. Investigate the effect of the physical properties by using other partially miscible liquid pairs over the maximum possible temperature range.

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- B. Develop techniques for determining dispersed-phase film coefficients and test equation (2) as a method of correlation.
1. Develop techniques for the preferential sampling of the dispersed phase in the presence of the continuous phase.
 2. Determine individual film coefficients for the unidirectional transfer of mass from the field phase to the dispersed phase.
 3. Using a third component (solute) determine the dispersed-phase film coefficient by assuming the additivity of film resistances and taking the difference between the measured over-all coefficient and the calculated continuous-phase coefficient.
 4. Determine the effect, if any, of the direction of mass transfer on the mass transfer coefficient by means of a three-component system.

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VII NOTATION

A	total interfacial area of dispersion in mixing vessel, sq cm
a	interfacial area per unit volume of total mixed phases, sq cm/cu cm
c	concentration of solute in extract-phase effluent, g/cu cm
c*	concentration of solute in extract phase at saturation, g/cu cm
D	impeller diameter, cm
D_v	molecular diffusivity of solute in extract phase, sq cm/sec
d	mean surface-volume drop diameter, cm
d_i	average drop diameter in size interval "i," cm
I_0/I	ratio of incident to emergent light intensities
K	over-all mass transfer coefficient cm/sec
k_c	individual mass transfer coefficient for the continuous-phase film, cm/sec
L	optical path length, cm
N	rate of mass transfer, g/sec
n	impeller speed, revolutions/sec
n_i	number of drops in size interval "i"
q_d	flow rate of dispersed-phase effluent, cu cm/sec
s, b, and h	constants
T	diameter of mixing vessel, cm
V_d	superficial velocity of dispersed-phase effluent based on vessel diameter, cm/sec
x_v	volume fraction of dispersed phase

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μ_f	viscosity of continuous phase, g/cm sec
ρ_f	density of continuous phase, g/cu cm
σ	interfacial tension, dynes/cm
N_{Ar}	area function, $\mu_f a/V_d \rho_f$
N_{Dr}	drop diameter function, $q_d \rho_f / \mu_f d$
N_{Re}	Reynolds number, $T V_d \rho_f / \mu_f$
$(N_{Re})_d$	Reynolds number with drop diameter as the characteristic length, $d V_d \rho_f / \mu_f$
N_{Sc}	Schmidt number, $\mu_f / \rho_f D_v$
N_{Sh}	Sherwood number, $k_c T / D_v$
$(N_{Sh})_d$	Sherwood number with drop diameter as the characteristic length, $k_c d / D_v$
N_{We}	Weber number, $T V_d^2 \rho_f / \sigma$

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APPENDIX**Graphs and Table of Supporting Data**

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FIGURE 8
REFRACTIVE INDEX OF ISOBUTANOL-WATER MIXTURES AT 25C

FIGURE 9
REFRACTIVE INDEX OF NITROBENZENE-ETHYLENE
GLYCOL MIXTURES AT 25C

FIGURE 10
OPTICAL DENSITY OF NITROBENZENE, ETHYLENE GLYCOL, AND WATER

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FIGURE 11
CORRELATION OF INTERFACIAL AREA WITH LIGHT TRANSMITTANCE FOR DISPERSIONS
OF ISOBUTANOL IN WATER

FIGURE 12
CORRELATION OF INTERFACIAL AREA WITH LIGHT TRANSMITTANCE FOR 25 PER CENT
(VOLUME) DISPERSIONS OF NITROBENZENE IN ETHYLENE GLYCOL

FIGURE 13

CORRELATION OF INTERFACIAL AREA WITH LIGHT TRANSMITTANCE FOR 10 PER CENT (VOLUME) DISPERSIONS OF NITROBENZENE IN ETHYLENE GLYCOL

FIGURE 15
 INTERFACIAL AREA AS A FUNCTION OF
 FLUID MOTION PARAMETERS

FIGURE 14
 EFFECT OF DISPERSED PHASE FLOW RATE
 ON INTERFACIAL AREA
 (Isobutanol-Water System in 4-in Vessel)

FIGURE 16
COMPARISON OF METHODS FOR THE CORRELATION OF MASS TRANSFER DATA

Table 4

PARAMETERS USED IN CORRELATIONS

Run	Surface- Volume Drop Diameter d_v (cm)	Drop Diameter Function N_{Dr} ($q_d \rho_f / \mu_f d$)	Area Function N_{Ar} ($\mu_f a / V_d \rho_f$)	Reynolds Number N_{Re} ($TV_d \rho_f / \mu_f$)	Drop Reynolds Number $(N_{Re})_d$ ($dV_d \rho_f / \mu_f$)	Sherwood Number N_{Sh} ($k_c T / D_v$)	Drop Sherwood Number $(N_{Sh})_d$ ($k_c d / D_v$)	Schmidt Number N_{Sc} ($\mu_f / \rho_f D_v$)
1A	0.0723	0.717×10^5	1.46	433	2.06	-	-	1470
2A	0.0331	2.35×10^5	2.12	650	1.41	-	-	1470
3A	0.0287	3.61×10^5	1.84	866	1.63	-	-	1470
4A	0.0273	4.55×10^5	1.61	1039	1.86	-	-	1470
5A	0.0239	6.07×10^5	1.58	1213	1.90	-	-	1470
6A	0.0233	6.67×10^5	1.50	1300	1.99	-	-	1470
13A	0.0227	6.85×10^5	1.54	1300	1.94	-	-	1470
14A	0.0237	6.56×10^5	1.48	1300	2.02	-	-	1470
15A	0.0323	2.41×10^5	0.972	974	3.09	-	-	1470
16A	0.0309	2.52×10^5	1.01	974	2.96	-	-	1470
17A	0.0316	3.28×10^5	0.743	1300	4.04	-	-	1470
18A	0.0228	5.68×10^5	0.824	1625	3.64	-	-	1470
19A	0.0202	7.69×10^5	0.773	1950	3.88	-	-	1470
20A	0.0181	2.86×10^5	1.30	650	1.16	-	-	1470
23A	0.0148	1.16×10^5	1.92	215	0.312	12.9×10^4	1.88×10^2	1470
24A	0.0152	1.13×10^5	1.87	215	0.321	7.87×10^4	1.18×10^2	1470
25A	0.0162	1.06×10^5	1.75	215	0.342	5.01×10^4	0.798×10^2	1470
26A	0.0197	2.63×10^5	1.19	650	1.26	14.5×10^4	2.81×10^2	1470
27A	0.0178	0.844×10^5	1.89	189	0.333	8.43×10^4	1.48×10^2	1470
28A	0.0215	1.51×10^5	1.39	406	0.858	10.9×10^4	2.30×10^2	1470
29A	0.0243	2.28×10^5	1.08	696	1.66	-	-	1470
30A	0.0260	3.31×10^5	0.869	1079	2.76	-	-	1470
1B	0.0371	5.44×10^3	11.1	25.5	0.0931	7.44×10^4	2.71×10^2	200,000
2B	0.0375	7.73×10^3	9.95	36.7	0.135	9.28×10^4	3.43×10^2	200,000
3B	0.0405	11.8×10^3	8.05	60.3	0.240	15.1×10^4	6.01×10^2	200,000
4B	0.0378	18.2×10^3	7.45	87.0	0.324	25.5×10^4	9.49×10^2	200,000
5B	0.0379	21.6×10^3	7.06	102	0.381	18.0×10^4	6.70×10^2	200,000
6B	0.0317	13.2×10^3	10.7	52.9	0.165	18.0×10^4	5.62×10^2	200,000
8B	0.0316	10.1×10^3	11.4	40.4	0.126	8.86×10^4	2.75×10^2	200,000
9B	0.0304	7.61×10^3	13.4	29.2	0.0873	6.77×10^4	2.03×10^2	200,000
10B	0.0357	6.07×10^3	11.5	27.3	0.0958	6.67×10^4	2.34×10^2	200,000
11B	0.0347	12.5×10^3	9.66	54.7	0.187	10.4×10^4	3.54×10^2	200,000
12B	0.0304	23.2×10^3	9.28	88.6	0.265	21.9×10^4	6.55×10^2	200,000
13B	0.0273	14.8×10^3	11.5	50.9	0.137	10.9×10^4	2.93×10^2	200,000
14B	0.0356	9.13×10^3	11.4	41.0	0.144	7.73×10^4	2.71×10^2	200,000
15B	0.0563	4.81×10^3	8.68	34.2	0.190	7.44×10^4	4.12×10^2	200,000
16B	0.0306	23.3×10^3	9.20	90.2	0.272	10.1×10^4	3.03×10^2	200,000
17B	0.0329	8.67×10^3	11.5	36.0	0.117	9.54×10^4	3.09×10^2	200,000
18B	0.0568	4.59×10^3	9.20	33.0	0.184	5.50×10^4	3.07×10^2	200,000

Runs 1A through 30A - isobutanol dispersed in water.

Runs 1B through 18B - nitrobenzene dispersed in ethylene glycol.

Runs 7A through 12A were omitted because of the presence of surface-active contamination in the isobutanol.

Run 7B data are not available because of the accidental breakage of the sampler.