Mass Transfer Studies Across Liquid/Liquid Interfaces (Use of an Analytical Ultracentrifuge)

Experimental data have been obtained using an Analytical Ultracentrifuge, in regions much closer to the interface in immiscible liquid/liquid systems than have hitherto been reported. The water/toluene system was investigated with acetic and propionic acids as the transfer solutes. The direction of mass transfer was from the water to the toluene phase. The data derived from the study indicates that the maximum mass transfer flux in the water/toluene system occurs, not at the interface, but at regions further away from it and that the water/toluene system offers significant interfacial resistance of about 10^4 s/cm to the transfer of acetic and propionic acids. The water/*n*-butanol system was also studied and found to offer negligible resistance to the transfer of acetic acid across the interface.

Diffusion coefficients for ethylene glycol, glycerol, and *n*-butanol in water were also determined with the Ultracentrifuge. The values derived in this study were reproducible to within 5% and were in close agreement with those derived from classical diffusion experiments.

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SCOPE

Mass transfer between two liquid phases is of importance in a variety of industrial operations and research activities. The role of the interfacial zone in the transfer process has been studied extensively, but much remains to be explained. Physical measurements near the interface disturb the system. Optical techniques have the advantage of providing information on the interfacial zone without disturbing the interface. Ward and Brooks (1952) and Davies and Wiggill (1960) used an optical technique to probe the interfacial region in an unstirred liquid/liquid system. Their data reveal very little information in regions within 1 mm from the interface.

This paper reports experimental data obtained with an Analytical Ultracentrifuge equipped with a schlieren optical system. These data are the first obtained by this technique and provide information on concentration gradients, in regions within 1 mm from the interface, in considerable detail.

Diffusion coefficients of ethylene glycol, glycerol, and *n*-butanol in water were determined with this instrument to check the results with those derived from classical diffusion experiments. The effect of centrifugal force on the diffusion process and the reliability and sensitivity of this optical technique were established before obtaining data on the immiscible liquid/liquid system.

The unstirred water/toluene system was investigated with acetic and propionic acids as the transfer solutes. Previous studies (Orell and Westwater, 1962) indicate that the interface in this type of system is disturbed during mass transfer by eruptions and spasms, while the regions close to the interface are agitated spontaneously. This phenomenon, known as *interfacial turbulence*, results in mass transfer rates several times higher than the ones predicted by the two-film theory (Blokker, 1957). Those studies were conducted under conditions where the deviations from the average temperature were at least ± 1°C. The equipment used in the present study permits temperature control in every run to within ± 0.1°C and temperature measurement to within ± 0.02°C. The objective of this study was to observe interfacial behavior under the isothermal conditions of the experiment and, particularly, to establish either the absence or presence of interfacial turbulence under these conditions.

CONCLUSIONS AND SIGNIFICANCE

The Ultracentrifuge was operated at a relatively low rotor speed of 6995 rev./min., and under these conditions the values of the diffusion coefficients were in close agreement with those derived by other investigators from experiments conducted under static conditions. Diffusion coefficients can be rapidly and accurately measured by this technique. Diffusion coefficients were measured at three relatively low concentration levels of 1.0, 3.0, and 5.0 wt. % and the values in this concentration range were constant to within ± 5%.

Since the values of diffusion coefficients measured in this study agreed well with those derived from experiments conducted under static conditions, it was established that the ultracentrifuge, when operated at the relatively low rotor speed, develops a centrifugal force just sufficient to produce sharp interfaces in immiscible liquid/liquid

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systems without significantly affecting the diffusion process. The reliability and sensitivity of the instrument were also established from the reproducibility of the data to within 5% in each of the experiments and the ability to control the temperature to within ± 0.1°C.

The maximum mass transfer flux in the water/toluene system (during the transfer of acetic and propionic acids across the interface) occurs, not at the interface, but at distances between 0.25 and 0.4 mm from the interface. The region between the plane of deflection of the schlieren pattern on the toluene side and the interface, which varies from about 0.25 to about 0.4 mm, depending upon the solute concentration, could represent the interphase between the water and toluene phases.

Using expressions derived from Crank's (1956) mathematical model to interpret the experimental data, it appears that the water/toluene system offers significant interfacial resistance to the transfer of solutes like acetic acid and propionic acids. At solute concentration levels of 6.9 and 10.0 wt. %, the interfacial resistances in the water/acetic acid/toluene system have been estimated to be in the range of 1.9×10^4 to 6.3×10^4 s/cm. In the water/propionic acid/toluene system, the interfacial resistance has been estimated to be 3.6×10^4 s/cm at a propionic acid level of 1.6 wt. %. These data substantiate results of a recent study (Blaschke and Schugerl, 1974) on a similar system using a liquid scintillation counting technique.

No interfacial turbulence was observed in any of the systems. This study revealed that the interface as well as the interfacial regions of the unstirred liquid/liquid system are quiet and motionless. The progress of the diffusion run was followed continuously by viewing the schlieren patterns through the screen viewer and any interfacial activity could have been easily detected with this sensitive instrument. The schlieren patterns (refractive index gradients) represent the concentration gradients of the solute in the diffusing medium at the low concentration levels used in this study. When the temperature control system was inactivated to permit temperature variations up to ± 1°C, the schlieren patterns were no longer well defined. This was probably due to disturbances in the interfacial region caused by local temperature gradients and could explain observations of interfacial turbulence and spontaneous emulsification made by earlier investigators using less sensitive temperature control systems.

Equilibrium was not established during the transfer process in the water/toluene system. This is clearly at variance with earlier studies (Ward and Quinn, 1965) on similar systems where interfacial equilibrium was observed to prevail at contact times of about ½ s and interfacial resistances were reported to be less than 100 s/cm.

The water/*n*-butanol system offered no significant resistance to the transfer of acetic acid across the interface. This is in agreement with the work of Fosberg and Heideger (1967) on a similar system. However, our schlieren data on this system reveals that equilibrium is not established during the course of the run which lasted more than 50 min.

A mathematical analysis of the nonequilibrium interface has been presented by Dang and Gill (1970). They report that the smaller the interfacial transfer coefficient, the larger is the time required for the interfacial concentrations to achieve their equilibrium values. This is not in agreement with the data presented in this study.

EXPERIMENT

Solvent Purities

The water used was doubly distilled. The glycerol, *n*-butanol, ethylene glycol, and toluene were of certified ACS grade. Glacial acetic acid was of analytical reagent grade and the propionic acid was obtained in purified form from National Biochemicals Corporation. No attempt was made to purify the reagents further.

Equipment

The Analytical Ultracentrifuge was a Model E, manufactured by Beckman Instruments, Inc. Presented in Figure 1 is a schematic of the internal parts of the Ultracentrifuge. Detailed information on the equipment is available in the Instruction Manual supplied by the manufacturer (Zeller, 1964). The equipment is incorporated with a schlieren optical system and a photographic unit to take pictures of the schlieren patterns at various time intervals. The temperature control unit regulates the constant temperature bath to within ±0.01°C.

Diffusion Cell

A view of the diffusion cell with the optical windows and the cell housing is shown in Figure 2.

The cell in this study is the one supplied by Beckman with the ultracentrifuge and is specially designed to contain the sample material and to permit light to pass through the sample during centrifugation. The cell has the following basic components: a centerpiece which contains the sample used for investigation, two windows made of optical glass that seal the open ends of the centerpiece, window holders that contain the windows, window liners and gaskets, and a cell housing that contains all the above-mentioned components. A screw ring firmly holds these components in the cell housing.

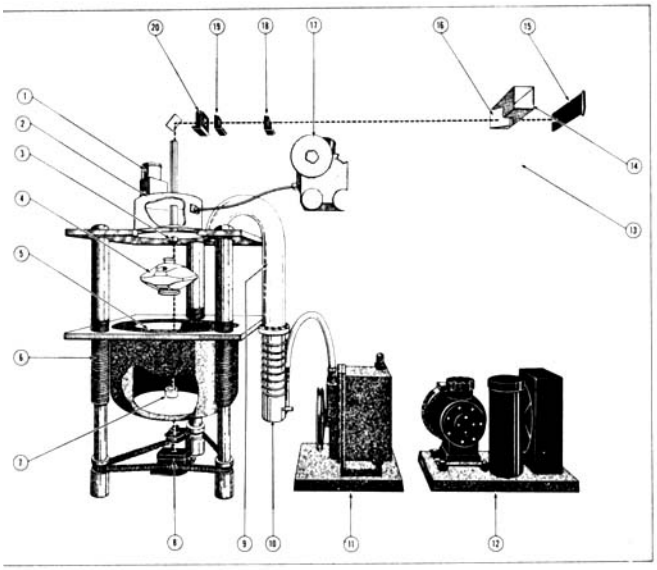
The centerpiece is the central component of the cell assembly. It contains the sample and is shaped to minimize convection. The thickness of the centerpiece represents the thickness of the fluid column through which light will pass. Choice of centerpiece thickness depends upon the difference in refractive index between the solutions under investigation and other factors such as the concentration of the solute, the degree of opacity, and the volume of sample used.

A valve-type synthetic boundary cell with an aluminum centerpiece (thickness: 12 mm) and a 4° sector angle was used in this study. The volume of sample that could be accommodated in the centerpiece was 0.83 ml.

In operation, the centerpiece is filled with about 0.4 ml of the denser solution, and the cup is filled with about 0.25 ml of the lighter solution. The maximum capacity of the cup is 0.3 ml. A hole in the centerpiece extends slightly into the sector cavity and accommodates the cup. In the shoulder of this hole is a smaller hole containing a cylindrical rubber valve. The valve is compressed against the bottom of the cup. The centrifugal force created at speeds between 3000 and 6000 rev./min, will compress the valve and then permit the solution in the cup to drain into the sector. The exact speed required depends on the material being studied. A boundary is now formed, and this marks the start of the diffusion experiment. A standpipe in the cup equalizes the pressure as the solution drains out of the cup. The rotor speed was maintained at 6995 rev./min. in all the experiments.

Experimental and Evaluation Procedures

The procedures involved in (1) preparing for a run, (2) assembling and filling the diffusion cell, (3) performing the run and (4) photographing the schlieren patterns during the course of the run, are outlined in the manufacturer's manual.



1	MOTOR HOUSING	11	VACUUM PUMP
2	DRIVE GEAR HOUSING	12	REFRIGERATOR
3	CONDENSING LENS	13	TEMPERATURE CONTROL UNIT
4	ROTOR	14	EYEPIECE
5	ROTOR CHAMBER (SHOWN IN OPEN POSITION)	15	PHOTOGRAPHIC PLATE
6	LIFT ROD	16	SWINGOUT MIRROR
7	COLLIMATING LENS	17	SPEED CONTROL GEARBOX
8	LIGHTSOURCE	18	CYLINDRICAL LENS
9	CONNECTING PIPE ROTOR CHAMBER TO VACUUM PUMP	19	CAMERA LENS
10	OIL DIFFUSION PUMP	20	PHASEPLATE

Fig. 1. Schematic perspective of the optical ultracentrifuge.

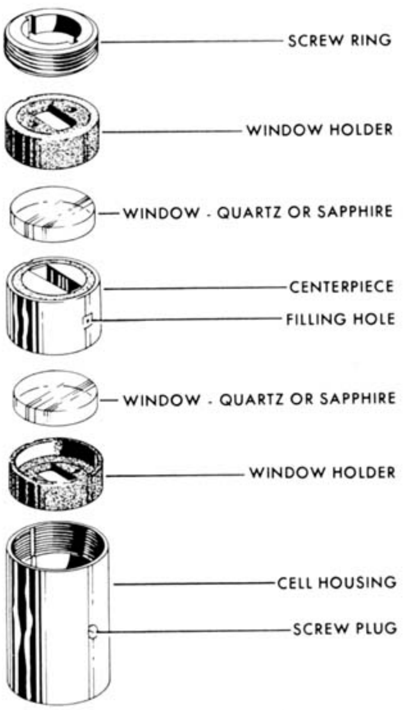


Fig. 2. Basic parts of the diffusion cell.

In the study of immiscible liquid-liquid systems, the aqueous and solvent phases were saturated with each other before the start of the diffusion run. In the n-butanol/water system, there is a separation of phases when the n-butanol concentration in the aqueous phase reaches 1 molar.

The schlieren patterns displayed in the viewing screen and/or the photographic plate represent the refractive index gradient distribution across the diffusion cell. The height of the schlieren pattern from the base line Y' is related to the refractive index gradient dn/dx by the following expression:

$$\frac{dn}{dx} = \frac{Y' \tan \theta}{La\, m_1 m_2} \tag{1}$$

where θ, L, a, m_1, m_2 are instrument constants.

The refractive index varies linearly with concentration in all the systems investigated in this study. This linear relationship makes it possible to obtain values for the concentration gradients across the cell directly from the schlieren patterns using the following relationship:

$$\frac{dc}{dx} = \frac{Y' \tan \theta}{La \dfrac{dn}{dc}\, m_1 m_2} = K\, Y' \tag{2}$$

where

$$K = \frac{\tan \theta}{La \dfrac{dn}{dc}\, m_1 m_2}$$

dn/dc may be either positive or negative. When it is negative in one phase and positive in the other, a reflection of the refractive index gradient curve will occur at the phase boundary (Linde, 1960).

Model Used for Interpretation of Data

The variation of concentration with time during diffusion may be found by integration of Fick's equation, applying the following boundary conditions for diffusion across an interface:

I.C. At $t = 0$, $x > 0$, $C = C_0$ and $x < 0$, $C = 0$

B.C. For $0 < t < \infty$, $x = 0$,

$$D_1 \frac{\partial C_1}{\partial x} + h\,(C_2 - C_1) = 0$$

$$D_1 \frac{\partial C_1}{\partial x} = D_2 \frac{\partial C_2}{\partial x}$$

Expressions for the concentration gradients were reduced from the solution given by Crank for the following cases:

1. Infinite two phase systems with negligible[*] interfacial resistance to mass transfer.

$$\left(\frac{\partial C_1}{\partial x}\right)_{x=0} = \frac{(D_2/D_1)^{1/2}}{1 + (D_2/D_1)^{1/2}}\, C_0 \left[\frac{1}{\sqrt{\pi D_1 t}}\right] \tag{3}$$

$$\left(\frac{\partial C_2}{\partial x}\right)_{x=0} = -\frac{1}{1 + (D_2/D_1)^{1/2}}\, C_0 \left[\frac{1}{\sqrt{\pi D_2 t}}\right] \tag{4}$$

2. Infinite two phase systems with significant[**] interfacial resistance to mass transfer.

$$\left(\frac{\partial C_1}{\partial x_2}\right)_{x=0} = \frac{(D_2/D_1)^{1/2}}{1 + (D_2/D_1)^{1/2}}\, C_0 h_1 \left[1 - \frac{2}{\sqrt{\pi}} \sqrt{D_1 t}\,\right] \tag{5}$$

$$\left(\frac{\partial C_2}{\partial x}\right)_{x=0} = \frac{1}{1 + (D_2/D_1)^{1/2}}\, C_0 h_2 \left[1 - \frac{2}{\sqrt{\pi}} h_2 \sqrt{D_2 t}\,\right] \tag{6}$$

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[*] Corresponds to $h_1 \sqrt{D_1 t} > 1.5$ and $h_2 \sqrt{D_2 t} > 1.5$.

[**] Corresponds to $h_1 \sqrt{D_1 t} < 1.0$, $h_2 \sqrt{D_2 t} < 1.0$.

Table 1. Experimental Diffusion Coefficients ($\times 10^5$ cm²/s)

Initial solute concn., C_0 wt. %	1.0			3.0			5.0		
Temp., °C	10	15	25	10	15	25	10	15	25
Glycol-water	1.0	1.30	1.50	1.04	1.26	1.47	1.03	1.30	1.50
Glycerol-water	0.6	0.85	0.98	0.65	0.86	1.11	0.62	0.83	1.07
n-Butanol-water	0.76	0.81	1.06	0.65	0.83	1.06	0.75	0.88	1.06

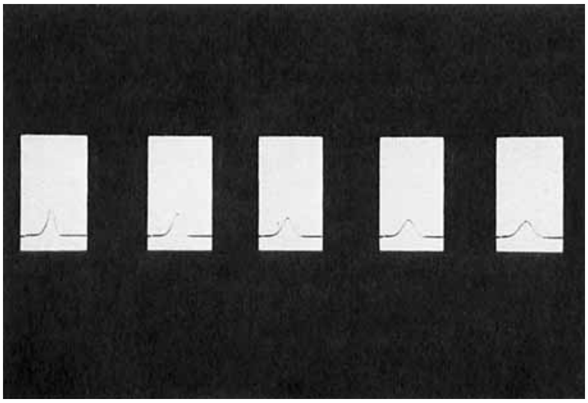

Fig. 3. Schlieren pattern—3% glycol diffusing into water. Temp. 15°C.

where h_1 and h_2 have been defined, for convenience, as follows:

$$h_1 = \frac{h}{D_1}\left\{ 1 + \left(\frac{D_1}{D_2}\right)^{1/2} \right\}$$

and

$$h_2 = \frac{h}{D_2}\left\{ 1 + \left(\frac{D_2}{D_1}\right)^{1/2} \right\}$$

Estimation of Diffusion Coefficients

The following expression for estimation of the diffusion coefficients was deduced from Equations (3) and (4) for $D_1 = D_2$.

$$\left(\frac{\partial C}{\partial x}\right)_{x=0} = \left[\frac{C_0}{2\sqrt{\pi D}}\right] \frac{1}{\sqrt{t}} \qquad (7)$$

RESULTS

Evaluation of Diffusion Coefficients

Typical schlieren photographs taken during the course of the diffusion experiments, for the glycol-water system, are presented in Figure 3. The duration of the experiment was 1830 s, with the first picture taken 270 s after the start of the run. The time interval between each picture is about 300 s.

The schlieren curves (concentration gradients) are well represented by Equation (7). Plots of Y' vs. $t^{-1/2}$ yield straight lines and the diffusion coefficients were evaluated from the slopes of these lines. A typical plot of Y' vs. $t^{-1/2}$ is shown in Figure 4.

The values of diffusion coefficients were reproducible to within 5% and, at the relatively low concentration levels, they were found to be independent of concentration. The diffusion coefficients evaluated in this study for the three systems, at three temperature and concentration levels, are presented in Table 1. Typical literature values are presented in Table 2 for comparison. The values derived from this study are in close agreement with published values.

Table 2. Typical Experimental Values of Diffusion Coefficients from Literature

System	Temp., °C	$D \times 10^5$ (cm²/s)
Glycol-water	25	1.15 ± 0.06
Glycerol-water	25	0.94 ± 0.06
	10	0.63 ± 0.07
n-Butanol-water	25	0.96 ± 0.05
	15	0.77 ± 0.07

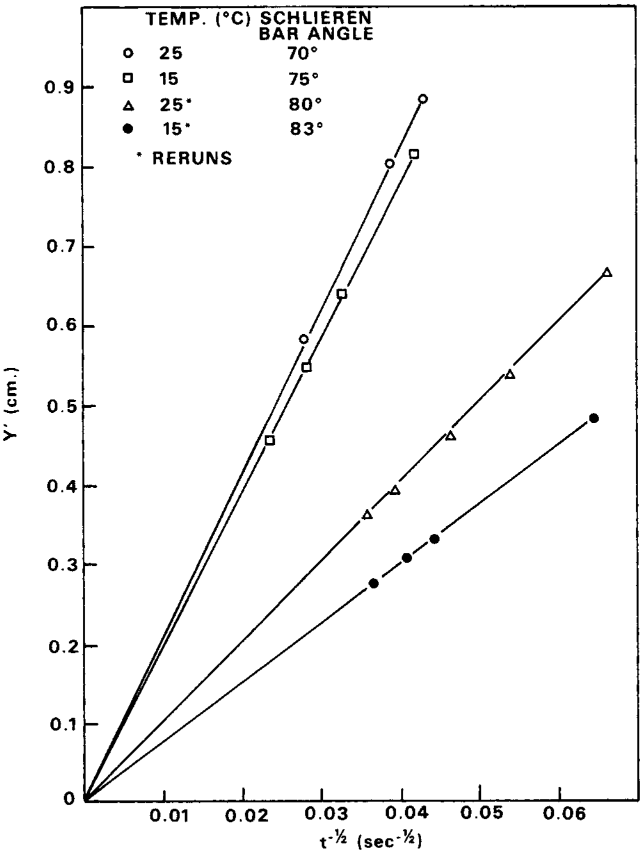

Fig. 4. 3.0% glycol solution-water.

Immiscible Liquid/Liquid Systems

Values of the diffusion coefficients of acetic and propionic acids in water and in toluene, reported by Ward and Brooks, were used in the analysis of our experimental data to estimate the interfacial coefficients in the water/acetic acid/toluene and water/propionic acid/toluene systems.

For analysis of the data obtained from the water/acetic acid/*n*-butanol system, the diffusion coefficients of acetic acid in water and in *n*-butanol were not readily available and were determined using the technique described in the previous section. The results are shown below:

Acetic acid in n-butanol, $D = 0.69 \times 10^{-6}$ cm^2/s

Acetic acid in water, $D = 0.85 \times 10^{-5}$ cm^2/s

1. *Water/Toluene System.* Magnified (10X) versions of the schlieren patterns for the water/acetic acid/toluene system, taken 5 and 14 min. after formation of the interface, are presented in Figures 5a and 5b. The interface is sharp and this makes it possible to get detailed concentration gradient information in regions within 1 mm from the liquid/liquid interface. When the heights of the schlieren peaks above the base line (Y_1, Y_2) at $x = 0$, on either side of the interface, are plotted against the square roots of the respective times ($t^{1/2}$), straight lines with negative slopes are obtained. When Y_1 is plotted against $t^{-1/2}$, there is a significant departure from linearity. This is shown in Figure 6. As indicated by Equations (5) and (6), this corresponds to the existence of significant resistance at the interfacial zone. The interfacial resistance was evaluated from slopes and intercepts of the lines in Figure 6.

The effect of solute concentration on the interfacial diffusion process was determined by raising the concentration of the acetic acid in the aqueous layer to 10.0 wt %. As in the previous case, the plot Y_1, Y_2 vs. $t^{1/2}$ yielded straight lines with negative slopes.

The diffusion behavior of propionic acid from the aqueous to the toluene layer in the water/propionic acid/toluene system was analogous to that of acetic acid.

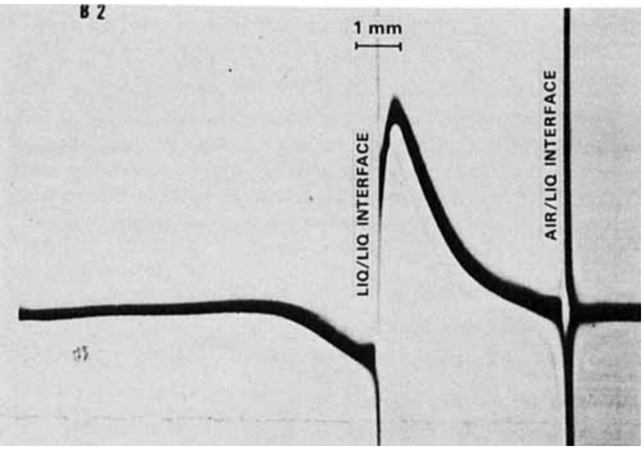

Fig. 5a. Schlieren pattern for water/toluene system. 5 min. after start of diffusion, $C_0 = 6.9\%$ wt %, Temp. 20°C, Magnification: 10.

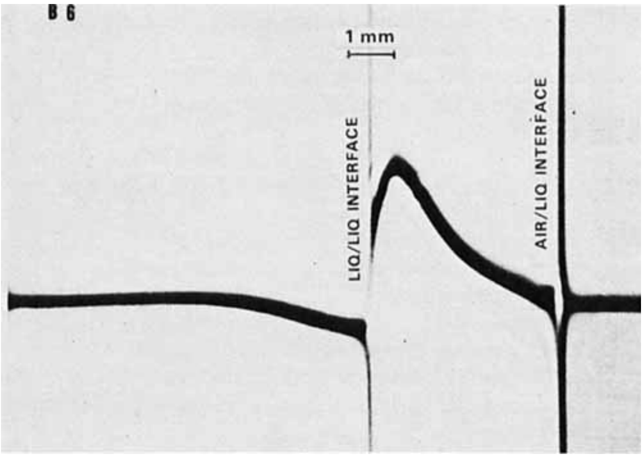

Fig. 5b. Schlieren pattern for water/toluene system. 14 min. after start of diffusion, $C_0 = 6.9$ wt %, Temp. 20°C, Magnification: 10.

Values of the interfacial transfer coefficients and resistances in the water/acetic acid/toluene system (at two concentration levels of acetic acid) and water/propionic acid/toluene system are summarized in Table 3.

2. *Water/n-Butanol System.* The initial solute (acetic acid) concentration in the aqueous layer was 2.04 wt %. The magnified version (10X) of the schlieren pattern, taken 50 min. after formation of the interface, is presented in Figure 7.

When the heights of the schlieren peaks above the base line (Y_1, Y_2) at $x = 0$ are plotted against $t^{-1/2}$, straight lines passing through the origin are obtained.[*] This is shown in Figure 8. The schlieren curves seem to be well represented by Equations (3) and (4), indicating that the interfacial zone offers negligible resistance to the transfer of the solute. When Y_2 is plotted against $t^{1/2}$, there is a significant departure from linearity.

VALIDITY OF DATA FOR IMMISCIBLE SYSTEMS

A procedure for checking the validity of both the data generated from this work and the model used for data

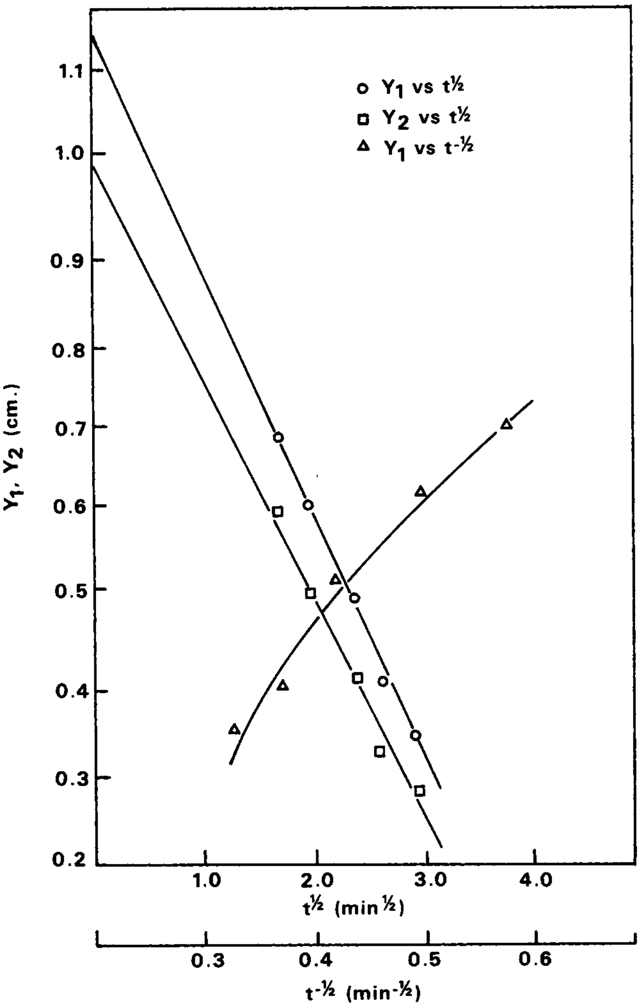

Fig 6. 6.9% acetic acid in water/toluene. Temp. 20 ± 0.1°C.

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[*] The zero-time could not be determined precisely in some of the experiments. This is reflected in the plot (shown in Figure 8) on which the lines intersect the abscissa instead of passing through the origin. The zero-time corrections in the various diffusion experiments conducted in this study varied from 0 to 40 s; and since the duration of an experiment was about 40 min., the zero-time correction is not considered significant.

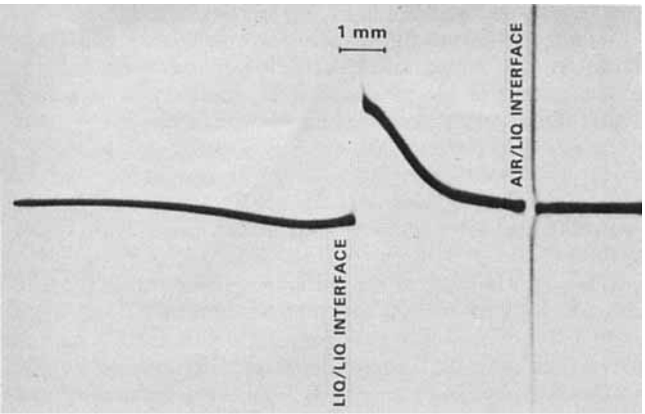

Fig. 7. Schlieren pattern for the water/n-butanol system, 50 min. after start of diffusion, $C_0 = 2.04$ wt %, Temp. 21.3°C, Magnification: 10.

TABLE 3. SYSTEM: WATER-TOLUENE
Temperature: 20 ± 0.1°C

Solute	Solute concn. in aq. phase, C_0, wt. %	Interfacial transfer coefficient $h \times 10^4$ cm/s	Interfacial transfer resistance $1/h \times 10^{-4}$, s/cm
Acetic acid	10.00	0.16	6.25
Acetic acid	6.90	0.53	1.89
Propionic acid	1.63	0.28	3.57

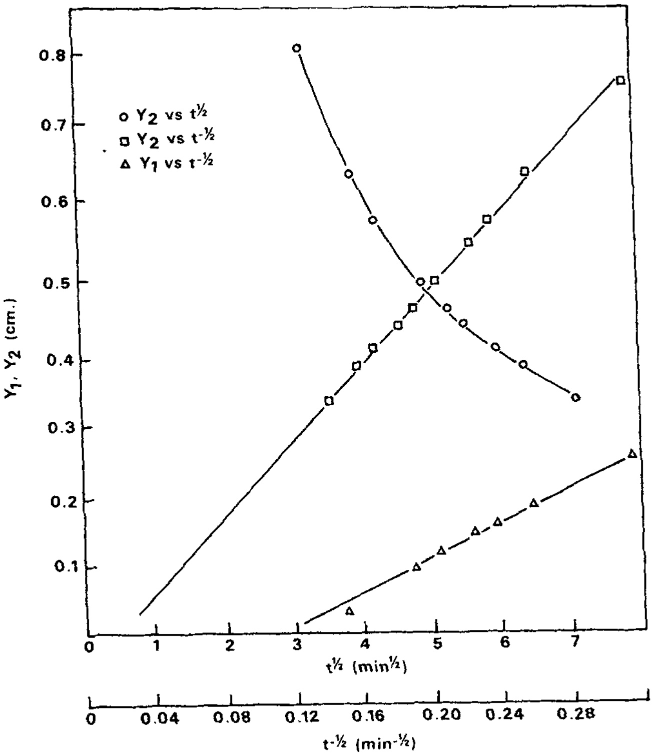

Fig. 8. 2.04% acetic acid in water/n-butanol Temp. 21.3 ± 0.1°C.

interpretation is outlined below. The data from the water/propionic acid/toluene system is used to illustrate the procedure.

Schlieren bar angle $\theta = 60°$

Initial solute concentration $(C_0) = 1.63$ wt. %

$$\left(\frac{dn}{dc}\right)_1 = 0.95 \times 10^{-3} \text{ (wt \%)}^{-1};$$

$$K_1 = \frac{\tan\theta}{La\left(\frac{dn}{dc}\right)_1 m_1 m_2} = 6.91$$

$$\left(\frac{dn}{dc}\right)_2 = 1.1 \times 10^{-3} \text{ (wt \%)}^{-1};$$

$$K_2 = \frac{\tan\theta}{La\left(\frac{dn}{dc}\right)_2 m_1 m_2} = 5.96$$

$$D_1 = 0.965 \times 10^{-5} \text{ cm}^2/\text{s}$$

$$D_2 = 1.774 \times 10^{-5} \text{ cm}^2/\text{s}$$

Substituting these values, Equations (5) and (6) reduce to

$$\frac{\partial C_1}{\partial x} = 1.04 \times 10^5 C_0 h - 5.73 \times 10^7 C_0 h^2 \sqrt{t} \quad (8)$$

$$\frac{\partial C_2}{\partial x} = 0.563 \times 10^5 C_0 h - 3.09 \times 10^7 C_0 h^2 \sqrt{t} \quad (9)$$

Ratio of slope to intercept from plot of Y_1, Y_2 vs. $t^{1/2} = 548\,h$

Estimated value of initial solute concentration from Equation (8) and intercept of Y_1 vs. $t^{1/2}$ plot $= 1.9$ wt %.

Estimated value of initial solute concentration from Equation (9) and intercept of Y_2 vs. $t^{1/2}$ plot $= 1.5$ wt %.

The agreement between estimated and actual values of solute concentrations is reasonably good, especially since values of D_1, D_2 $(dn/dc)_1$, and $(dn/dc)_2$ were taken from different sources and had to be derived by interpolation.

ACKNOWLEDGMENTS

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NOTATION

a = thickness of fluid column along optic axis
C = solute concentration, wt. %
D = solute diffusion coefficient, cm²/s
h = interfacial transfer coefficient, cm/s
h_1, h_2 as defined
L = optical lever arm, distance from the upper collimating lens to the plane of the Schlieren diaphragm
m_1 = magnification factor for camera lens
m_2 = magnification factor for cylindrical lens
n = refractive index
t = time
X = coordinate normal to interface
Y' = height of Schlieren peak at the interface $(x = 0)$ above the base line for the miscible liquid/liquid systems
Y_1, Y_2 = height of Schlieren peaks above base line (at $x = 0$) for immiscible liquid/liquid systems
θ = angle of inclination of Schlieren diaphragm

Subscripts

0 = initial condition
1 = aqueous phase
2 = organic phase

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Estimation of Bubble Diameter in Gaseous Fluidized Beds

Bubble size is one of the most important parameters in the design and simulation of a fluidized-bed reactor.

A correlation of the bubble size and growth in fluidized beds of various diameters is developed. A maximum bubble diameter determined from the bubble coalescence is incorporated in the correlation to relate the effect of the bed diameter on the bubble size.

Experimental data of bubble size reported are used to develop and test the validity of the correlation. The bubble diameters calculated using this correlation show good agreement with the observed bubble diameters.

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SCOPE

A correlation of bubble diameter for fluidized beds of various sizes including pilot scale is presented.

In recent years a number of fluidized-bed models (Mori and Muchi, 1972; Kato and Wen, 1969; Kunii and Levenspiel, 1968; Toor and Calderbank, 1967; Partridge and Rowe, 1966; Kobayashi and Arai, 1965; Orcutt et al., 1962) which take into consideration the behavior of bubbles have been proposed for predicting the performance of fluidized-bed reactors. Ishida and Wen (1973), Wen (1972), and Yoshida and Wen (1970) have also pointed out the importance of the bubble behavior in the coal conversion processes such as gasification and combustion.

In these studies one of the most important factors governing the extent of chemical conversion is the diameter of bubbles in the bed.

Although many correlations for estimation of the bubble diameter in fluidized beds (Yasui et al., 1958; Kato and Wen, 1969; Park et al., 1969; Whitehead et al., 1967; Rowe et al., 1972; Geldart, 1971; Chiba et al., 1973) are available, none of these correlations can predict the effect of the bed diameter on the bubble diameter.

In this paper, the bubble size and bubble growth rate are examined in light of the bed diameter and the design of distributor plates. A semi-empirical equation for bubble growth in fluidized beds of various sizes including pilot-plant scale is presented.

CONCLUSIONS AND SIGNIFICANCE

A correlation which predicts bubble diameters in freely bubbling fluidized beds and which accounts for the effect of the bed diameter on the bubble diameter is presented.

The proposed bubble growth correlation has the form

$$\frac{D_{BM} - D_B}{D_{BM} - D_{B0}} = \exp(-0.3h/D_t)$$

where D_B is the diameter of the bubble, D_t is the bed diameter, and h is the elevation or the height above the distributor plate. Initial bubble diameter formed at the surface of the perforated plate is calculated from

$$D_{B0} = 0.347\{A_t(u_0 - u_{mf})/n_d\}^{2/5}$$

where A_t is the cross-sectional area of the bed, u_0 is the