

MASS TRANSFER IN A STIRRED TRANSFER CELL WITH A FLAT INTERFACE

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Abstract—A agitated vessel of Lewis cell type was used to investigate the effect of physical properties on the mass transfer coefficient for partially miscible binary systems. Some measurements were performed with ternary systems transferring only one solute across two immiscible solvents. The mass transfer coefficients were measured under the conventional contra-rotating conditions which were behaved as if the interface was not rotating for some combinations of agitation speeds in each of the two phases.

The mass transfer coefficient was deduced from solving a steady-state two dimensional convective-diffusion equation with the assumption of sinusoidal motion of eddies. Owing to the complexity of the hydrodynamic conditions near the liquid-liquid interface, theoretical approach was impossible. Thus, the effects of forced turbulence and physical properties on the effective surface renewal time were experimentally investigated. The relation between the mass transfer coefficients and the relevant variables was obtained by conventional dimensional analysis as follow:

$$Sh_w = 1.93 \times 10^{-3} Ca^{1/2} Sc_w^{0.5} Re_w^{0.70} \exp(1.60 \times 10^{-4} \frac{\nu_w}{\nu_o} Re_o)$$

INTRODUCTION

It is necessary to understand the mechanism of diffusion across a liquid-fluid interface because it is very important to kinetic analysis of solvent extraction with chemical reaction and various chemical processes. But the mechanism of mass transfer through free interface has been not fully understood. Specially because of important property of the free interface, i.e. its mobility, it can be assumed that there is the exchange of momentum between both sides of the interface.

The stirred transfer cell originally used by Lewis [1] is a convenient apparatus for studying the mechanism of mass transfer across an interface of known area between two turbulent liquid phases. Several studies [1-5] have been made on the correlations for the mass transfer coefficients in agitated vessels with a flat interface. However, there has been a lack of agreement about the influence of physical properties on the mass transfer rate in their correlations.

The aim of this work is to study the mechanism of mass transfer and the effects of physical properties on the mass transfer rate for binary systems and ternary systems.

THEORETICAL BACKGROUND

In earlier models such as film model, penetration model and surface renewal model, the fluid elements near the interface were thought to be either solid or laminar. Scriven [6] pointed, however, out that the convective effects play a major role in affecting the diffusion transport in the interfacial zone, and later models included convection terms.

In a stirred tank, the secondary radial and axial circulation patterns are superimposed on the primary rotational motion generated by stirrer. The primary circulation flow and the secondary circulation flow will collide with each other and numerous eddies of different size and strength may be generated by collision. Since the primary circulation flow is predominant in the stirred tank with an impeller, the average tangential velocity is much higher than the axial or radial velocity at a fixed location near the interface. The average tangential velocity will fluctuate around a mean value because of the continuous collision of the secondary circulation flow on the primary circulation. In this work, the convective effect of turbulence on the flow field near the interface was represented by eddy sweeping fresh liquid to the vicinity of the interface. Since the tangential velocity is dominant over the

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radial velocity, the flow can be considered approximately two-dimensional at a fixed radial position. In general, scales of eddy size considerably smaller than the dimension of the vessel are assumed to exist, and for such scale the surface appears nearly flat.

The eddy rises up from the bulk, comes into contact with the interface, then goes back down to the bulk. The main features of the upward motion may be well represented by two-dimensional form of a sinusoidal motion (see Fig. 1) as:

$$u = V_m \sin\left(\frac{\pi x}{L}\right) \tag{1}$$

where V_m is the maximum x-component velocity and L is the characteristic eddy length in the x-direction.

For solving a steady-state two dimensional convective diffusion equation, assumptions are as follows:

- 1) The x-direction diffusion is neglected because the concentration gradient in the x-direction is small.
- 2) The depth of penetration by diffusion is smaller than the thickness of the element of fluid in contact with the interface.

- 3) Flow is incompressible, the diffusivity is independent on concentration, and the diffusion-induced interfacial velocity is negligible.

The convection-diffusion equation can be simplified by using the assumptions and equation of continuity:

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \tag{2}$$

$$v = - \int \left(\frac{\partial u}{\partial x}\right) dy = -y \frac{du}{dx} \tag{3}$$

The resulting simplified equation is:

$$V_m \sin\left(\frac{\pi x}{L}\right) \frac{\partial C}{\partial x} - \frac{\pi V_m y}{L} \cos\left(\frac{\pi x}{L}\right) \frac{\partial C}{\partial y} = D \frac{\partial^2 C}{\partial y^2} \tag{4}$$

The corresponding boundary conditions are:

$$C = C_b \quad \text{for } x = 0 \tag{5a}$$

$$C = C_s \quad \text{for } y = 0 \tag{5b}$$

$$C = C_b \quad \text{for } y \rightarrow \infty \tag{5c}$$

where C_b and C_s are the bulk and the interface solute concentration, respectively.

The general solution for the Eq. (4) with boundary conditions, Eqs (5), can be given by using the similarity variable transformation. The solution is expressed as [7,8]:

$$\frac{C - C_b}{C_s - C_b} = \operatorname{erfc} \left\{ \frac{y}{2} \sqrt{\frac{\pi V_m}{DL} \left(1 + \cos \frac{\pi x}{L}\right)} \right\} \tag{6}$$

The local mass transfer, k_L , of the single eddy can be obtained from the slope of the concentration profile at $y=0$, and the average mass transfer coefficient, k , is

$$k \propto \sqrt{D \frac{V_m}{L}} \tag{7}$$

The essential difficulty in the eddy models is with the arbitrary and conflicting assumptions regarding the group of motions controlling the transfer process. According to Levich [9], the characteristic eddy velocity, V , is utilized to characterize the mass transfer properties of the interface. For the small-eddy model, the rate of dissipation, ϵ , is needed instead. According to the large-eddy model, the turbulence intensity, V , and macroscale, L , are the relevant turbulence properties. In this work, the characteristic time L/V_m may also be viewed as effective renewal time for the motions in the vicinity of the interface.

EXPERIMENTAL

1. Apparatus

The experiments in this work were carried out in a

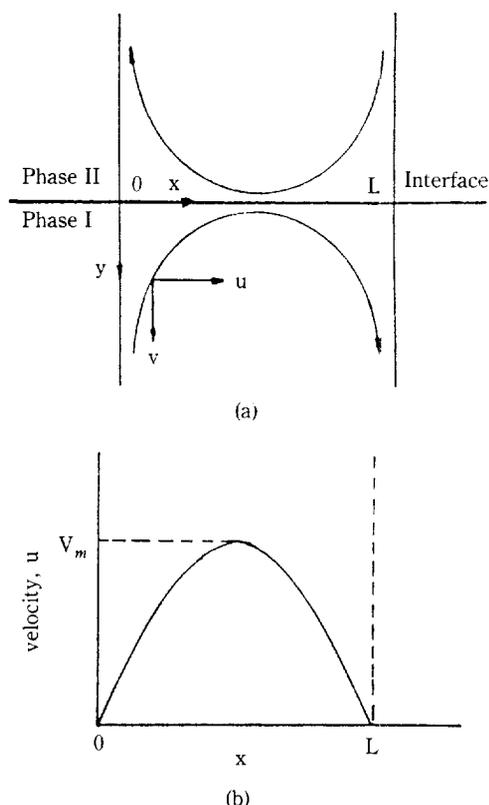


Fig. 1. (a) Coordinate system used in eddy analysis
(b) x-component velocity close to interface

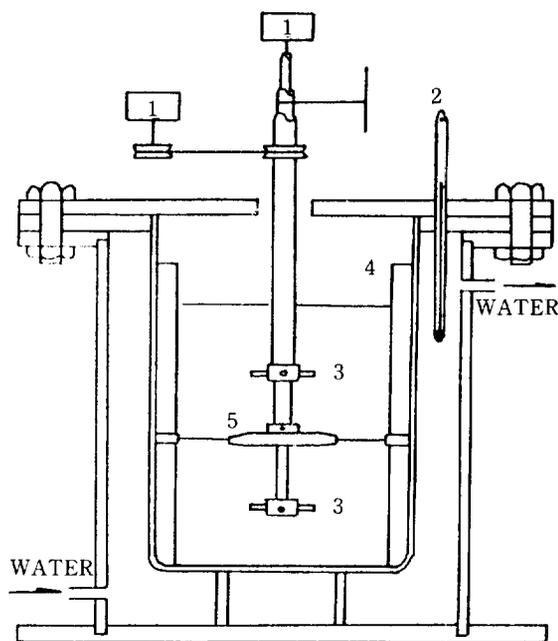


Fig. 2. Experimental apparatus.

1. motor
2. thermometer
3. impeller
4. vertical baffle
5. central circular baffle

stirred transfer cell (Fig. 2) with impellers in both phases. The vessel consisted of an inner glass cylinder 120 mm. in diameter and 150 mm. in height. The annular space between inner and outer cylinders was used as a water jacket.

The two glass compartments were separated by a circumferential baffle and a central circular baffle. Four equally-spaced vertical baffles, one-tenth of the vessel diameter in width, were attached to the internal wall of the vessel, and these together with outer circumferential baffle. In this work interface was restricted an annular gap between the central circular baffle preventing vortex and the outer baffle reducing irregular wall effects.

In both phases, each stirrer was a paddle agitator with four blades, having the diameter of four-tenths of the vessel diameter and the height of one-tenth of the each liquid phase height respectively. The two stirrers were placed at the midpoint of each liquid phase, and independently driven by variable speed d.c. motors. All metal parts contacting with the liquids were made of stainless steel.

2. Systems

The majority of experiments was carried out with the partially miscible binary liquid systems. According to the phase rule, such systems have no degrees of freedom at constant temperature. Therefore, partially miscible binary systems should be free of interfacial-induced instability. Hence, such binary systems are very interest in the investigation of liquid-liquid mass transfer and could be used to produce reference mass transfer correlations. Some measurements were performed with the ternary systems transferring only one solute between two immiscible solvents.

Physical properties of the chemicals used are shown in Table 1. Values of the diffusivity were calculated by the formula of Sitaraman et al. [10]. Commercial preparations of organics rated as extra pure grade reagent were employed.

Table 1. Physical properties of systems used.

Physical Property	Binary systems with water			Ternary systems		
	ethyl acetate	n-butanol	iso-butanol	water	benzene	CCl ₄
Density (Kg/m ³)	891	804	796	997	862	1582
Viscosity × 10 ⁻² (Kg/m.s)	0.54	2.10	2.74	0.895	0.67	1.03
Diffusivity in water ¹ (m ² /s) × 10 ⁹	1.00	0.90	0.94	1.12		
Diffusivity in organic ² (m ² /s) × 10 ⁹	3.60	0.28	0.53			1.86
Interfacial tension (N/m) × 10 ³	5.5	2.5	2.1		28.7*	45.0*

1 of organic in binary systems; of acetone in ternary systems

2 of water in binary systems; of acetone in ternary systems

*: pure solvent

The concentrations of water in organic, in the binary experimental run, were determined by Karl-Fisher titration method while the contents of the organic solutes in the water phase were analyzed by refractometry. Analysis of refractometry was made by measurement of the refractive index using refractive index vs. concentration graphs which had been experimentally determined beforehand. The relation between the refractive index and concentration of various organics in the aqueous solution was almost linear in the range of the present measurement.

3. Procedure

Before each run the vessel and stirrer unit were thoroughly cleaned with chromic acid and distilled water. The measurements were started by filling the cell first with the heavier phase and then with the lighter one. After turning on the stirrers, samples were regularly taken by a syringe for analysis from both phases at 5-10 min. intervals. The stirring rates necessary to give the correct Reynolds numbers had been previously calculated and the controllers set at this speed. During each run the stirrer rates were frequently checked.

All experiments were carried out at $25 \pm 0.2^\circ\text{C}$ in batch operation. The mass transfer coefficients were measured under the conventional contra-rotating conditions in the all experiments. For the binary experiments, both phases were initially unsaturated. But for the ternary systems, solvents in both phases were saturated with each other beforehand.

RESULTS AND DISCUSSION

1. Mass Transfer Coefficients

If it is assumed that equilibrium exists at the interface in the binary systems, then the interfacial concentration is constant and substantially equal to the saturation value. Therefore the mass transfer coefficient for the binary systems is given by:

$$\ln |W_i^* - W_i| = -k_i \rho_i At / G_i + B \tag{8}$$

where W_i^* is equilibrium concentration of the solute and B is integration constant.

For the ternary systems, the experimental concentration range was chosen so as to permit linear approximation of the equilibrium curve, given by

$$W_w^* = mW_o + q \tag{9}$$

The concentrations on either side of the interface are normally assumed to be in equilibrium. Since the interfacial concentrations cannot be determined, it is customary to define an overall mass transfer coefficient based on the assumption that the resistance to

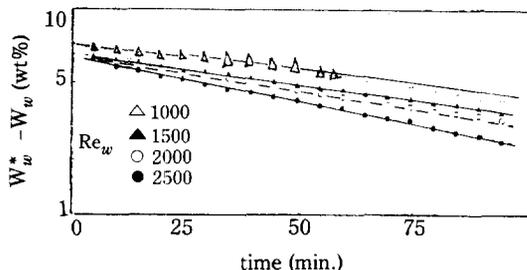


Fig. 3. Plot of concentration driving force vs. time in water phase for water-n-butanol system ($Re_w = 1000$).

transfer lies entirely in one phase. Then at any time the mass transfer coefficient based on the water phase is given by

$$\begin{aligned} \ln |W_w - \beta| &= -\alpha K_w t + B \\ \alpha &= (G_o + mG_w) A \rho_w / G_o G_w \\ \beta &= G_o (mG_w W_w^o / G_o + mW_o^o + q) / (G_o + mG_w). \end{aligned} \tag{10}$$

From each run in the cell, the concentration-time curves for the both phases were produced. For the water-n-butanol system, typical mass fraction driving force-time curves were given in Figs. 3 and 4. In all cases, good straight lines were obtained by a semi-log plot of $\ln |W^* - W|$ against t , and mass transfer coefficients, k , were obtained from the slope of the line of best fit relating $\ln |W^* - W|$ with t by least-square method.

The k_w/k_o ratios were 1.87 ± 0.11 for water-n-butanol system, 1.40 ± 0.09 for water-iso-butanol system and 0.53 ± 0.03 for water-ethyl acetate system without regard to the ratio of Reynolds number of both phases. These values were in accordance with 1.82, 1.64 and 0.60 proposed by Bulicka and Prochazka [4], respectively.

2. Effects of Diffusivity

If it is assumed that the frequency of the renewal in both layer is the same, from Eq. (7)

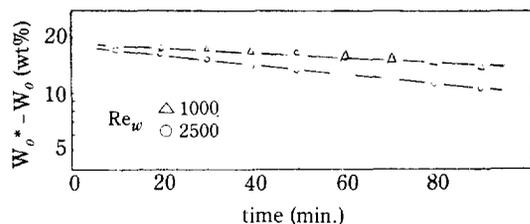


Fig. 4. Plot of concentration driving force vs. time in organic phase for water-n-butanol system ($Re_w = 1000$).

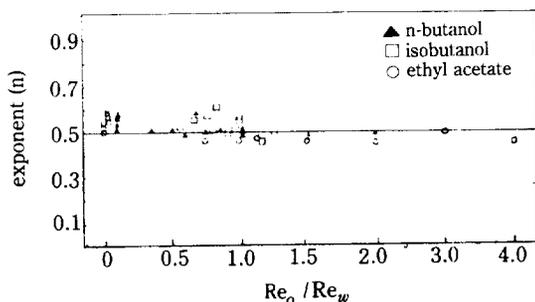


Fig. 5. Plot of n vs. Re_o/Re_w for binary systems.

$$\frac{k_w}{k_o} = \left(\frac{D_w}{D_o}\right)^{1/2} \quad (11)$$

Usually it has been postulated that k was proportional to D^n , with the same hydrodynamic conditions [3]. Fig. 5 is a plot of exponent, n , against the ratio of aqueous phase Reynolds number to organic phase. For the binary systems n was between 0.60 and 0.45; the mean was 0.52 with a standard deviation of 0.04, in good agreement with the value, 0.5, obtained from Eq. (11).

Surface renewal conditions exist at the interface as far as forced turbulence reaches the interface, and a transition to laminar boundary layers at the interface occurs as the intensity of agitation is reduced. In this work it was confirmed that the mass transfer coefficient was related to $D^{0.5}$ for higher agitation and showed that the exponent on D was between 0.5 and 0.67 in the transition regime between surface renewal and laminar boundary layer conditions for lower stirrer speeds.

In terms of other theoretical models the value of n can be induced from model based on a laminar boundary layer, for which $n=2/3$. And penetration and surface renewal theories which predict $n=0.5$ are applicable. McManamey et al. [3] based their correlation on the Levich model and Bulicka and Prochazka [4] based theirs on the surface renewal model, involve the dependence of k_w on $D^{1/2}$ as a natural consequence of their models. The other previous empirical correlations [1, 2] resulted in the effect of D varying with the power of 0 to 0.37, which has no plausible theoretical basis.

3. Effects of Reynolds numbers

The binary systems were studied within a range of Reynolds numbers before the interface was disrupted. On plotting the coefficients against Reynolds numbers on logarithmic coordinates using the Reynolds number of the other phase as a parameter a family of smooth curves were obtained (Fig. 6). It was great dependence of k_w on Re_w at small values of Re_o , but

the effect of Re_w diminished with increasing Re_o .

The binary systems and the ternary systems are shown various degrees of instability based on the work of Ying and Sawistowski [12]. However regardless of the degree of instability, it can be seen that k_w was proportional to about 0.7 power of Re_w when there is no agitation in the organic phase. The exponent of Re_w by this work was significantly smaller than that of 3/2 by McManamey et al. [3], and was comparable to 3/4 by Bulicka and Prochazka [4].

4. Correlation for Binary Systems

It is very interested in to investigate the liquid-liquid mass transfer in partially miscible binary systems. With the realization of the influence of the Marangoni effect on mass transfer rates, the advantages of binary systems become even more obvious. A constant interfacial composition implies a constant interfacial tension which would remain unchanged even in the case of surface renewal under the normal assumption of instantaneous attainment of equilibrium at the interface.

Unfortunately there is not a great deal of direct experimental information relating to the velocity structure of turbulent flows near a free interface. Besides, owing to the complexity of the hydrodynamic conditions near the liquid-liquid interface by contra-rotating, no theoretical approach is possible. Thus, in this work the relation between effective renewal time and relevant variable based on bulk phase was obtained by

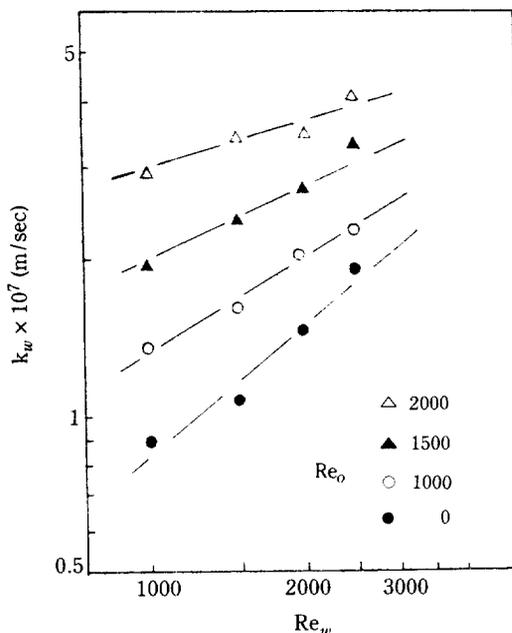


Fig. 6. Effect of Re_w and Re_o on mass transfer coefficient for water-n-butanol system.

conventional dimensional analysis. During the course of a run, the bulk concentration was an exponentially increasing function of time. In general, the physical properties are strongly dependent on concentration. In this work, however, it was used the physical properties of pure solvents for correlation of experimental data. Of course, to obtain more accuracy relation, it seems that the physical properties should correspond to the interfacial composition.

Fig. 6 indicates that Re_w and Re_o cannot be combined in the form of a product. However, using two contra-rotating stirrer assemblies in water phase, Davies and Khan [13] could adjust rotating speeds so that a whole is non-rotating, and led to the following empirical relation for surface renewal rate, s ,

$$\ln s = N/a - b \quad (12)$$

where N is expressed in revolution per minute.

In this study at certain combination of contra-rotating of the impellers, the interface was behaved as if a whole was absolutely static. Assuming the interaction of Re_w and Re_o on the water phase Sherwood number, Sh_w , the type of expression of Davies and Khan [13] was used for correlation of the experimental data. All binary systems were correlated by following eqn.

$$Sh_w = C_1 Sc_w^{C_2} Re_w^{C_3} \exp(C_4 \frac{\nu_w}{\nu_o} Re_o) \quad (13-1)$$

The coefficients in Eq. (13-1) were calculated from the experimental data and the physical properties of the aqueous phase, with an IBM 370 computer and statistical analysis system (SAS) of Barr et al. [14] to fit and evaluate the data statistical.

$$Sh_w = 0.116 Sc_w^{0.5} Re_w^{0.70} \exp(1.60 \times 10^{-4} \frac{\nu_w}{\nu_o} Re_o) \quad (13-2)$$

Fig. 7 shows a comparison of the values of Sh_w obtained in the present work with those calculated from Eq. (13-2), with an average deviation of 10.5% and regression coefficient of 0.985.

5. Application of the Binary Data to the Ternary Systems

The ternary systems were chosen for investigating the applicability of the binary data to the ternary systems. According to the phase rule, partially miscible binary systems have no degrees of freedom at constant temperature so that under isothermal conditions their interfacial concentrations are given by the corresponding solubility values. The driving force for mass transfer in each phase is thus the difference between the solubility and the bulk concentration, which is usually equated to the flow-average concentration. Since the interfacial concentrations cannot be determined in ternary systems, it is customary to define an

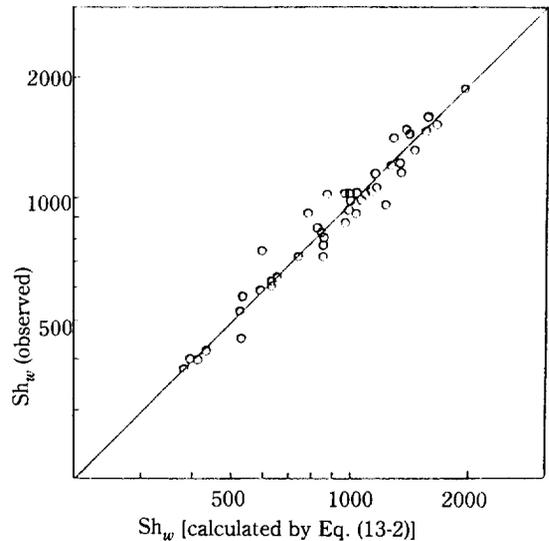


Fig. 7. Comparison of observed and calculated Sh_w for binary systems.

overall mass transfer coefficient based on the assumption that the resistance to transfer lies entirely in one phase. And the individual mass transfer coefficient of binary system is preferable to the single overall coefficient normally obtained in the transfer of a solute between two immiscible solvents.

It has been well known that mass transfer coefficients were influenced by the concentration driving force [15] as well as Marangoni instability [16].

With the linear distribution overall mass transfer coefficient based on water phase is given by

$$\frac{1}{K_w} = \frac{1}{k_w} + \frac{m}{k_o} \quad (14)$$

and as such is independent of concentration and concentration driving force as long as the system does not exhibit interfacial instability. Transport of mass is nearly always accompanied by local density and interfacial tension gradients, and it has been shown that mass transfer between two stationary phases or laminar films may be significantly affected by convection induced by such gradients [17]. However, with forced turbulence reaching as far as the interface one can expect that this effect would be unimportant.

Similarly with binary systems, overall mass transfer coefficients based on the water phase of ternary systems were calculated from the relation of Eq. (10). Fig. 8 shows a plot of overall mass transfer coefficients against Reynolds number of the water phase on logarithmic coordinate using the Reynolds number of the organic phase as a parameter.

It seemed that ternary mass transfer data failed to

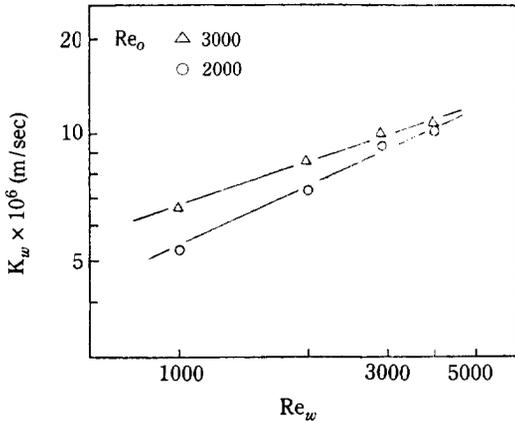


Fig. 8. Effect of Re_w and Re_o on K_w for water-acetone-benzene.

fit in Eq. (13-2) because the ternary data were too small to correlate with binary data. The greatest difference between the binary systems and the ternary systems is the viscosity of the organic phases and interfacial tension with organic phase. But in binary systems it was seemed that viscosity of organic phase had no effect on the mass transfer rate.

It has usually been assumed that interfacial tension did not affect liquid-liquid mass transfer rate. However basic prerequisite of Levich's theory [9], as well as that of Davies [11], is the decay of the vertical velocity component against interface within the zone of damped turbulence, while the velocity component along the interface is assumed to remain constant. The thickness of the zone is assumed from a balance of the inertial and surface tension forces, and also for a horizontal interface the gravity force. This model based on the concept that approach of an eddy to the interface is restrained by interfacial tension and gravitational forces. In the developed models from this, the influence of interfacial tension is included in $\sigma^{-0.5}$.

Asai et al. [5] have been shown that K_w was proportional approximately to -0.5 power of interfacial tension over a variation of σ of about 12 times.

The effect of interfacial tension was introduced to correlate the mass transfer data of ternary systems. When both interfacial tension and gravitational pressure are resisting the eddy thrust pressure, Davies [11] shows the following eqn from the pressure balance on eddy

$$\sigma_{eq} = \sigma_l + l^2 \Delta \rho g / 16 \quad (15)$$

where l is related to the scale of turbulence, which, for eddies produced by an impeller, is some fraction of impeller length and does not vary with speed. When the eddies are relatively large while the interfacial tension

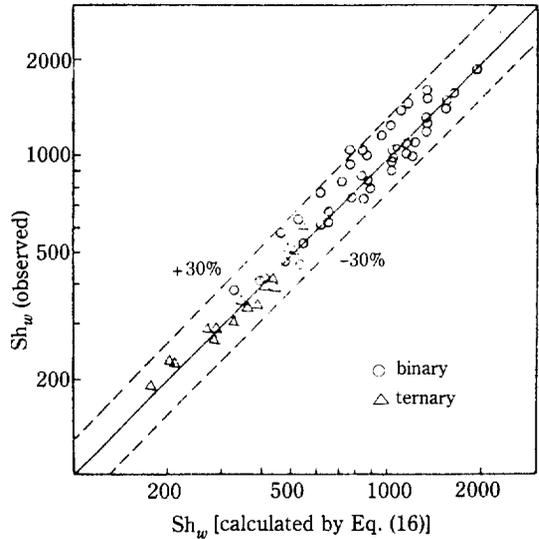


Fig. 9. Comparison of observed and calculated Sh_w for binary and ternary systems.

is relatively low, equivalent interfacial tension will be equal hydrostatic pressure. For smaller eddies, the interfacial tension term is dominant at a horizontal surface. All systems were correlated satisfactorily by following eqn.

$$Sh_w = 1.93 \times 10^{-3} Ca^{1/2} Sc_w^{0.5} Re_w^{0.70} \exp(1.60 \times 10^{-4} \frac{\nu_w}{\nu_o} Re_o) \quad (16)$$

Here the constant, 1.93×10^{-3} , is the proportionality constant characterizing the cell geometry, and is independent of the impeller speed and the liquid systems used.

Fig. 9 shows a comparison of the values of Sh_w obtained in the present work with those calculated from Eq. (16), and the observed values of Sh_w are in reasonable agreement with the calculated ones, with a maximum deviation of about 30 per cent.

CONCLUSIONS

A steady-state two dimensional convection diffusion equation was applied for predicting the mass transfer coefficient, and rates of mass transfer of the binary systems and the ternary systems were studied under conditions of contra-rotating. The effect of physical properties and turbulence intensity on effective surface renewal time was investigated. And measured mass transfer coefficients could be correlated with reasonable accuracy by Eq. (16). This correlation is useful regardless of the degree of instability.

NOMENCLATURE

a, b : constant in eq. (12)
 A : area of interface, m²
 B, B' : integration constant's in eqns (8) and (10)
 C : concentration, Kg/m³
 C₁₋₅ : coefficients
 Ca : Capillary number, $g d^2 \rho / \sigma_{eq}$
 d : diameter of impeller, m
 D : diffusion coefficient, m²/s
 g : gravitational constant, m²/s
 G : mass of phase, Kg
 k : individual mass transfer coefficient, m/s
 K : overall mass transfer coefficient, m/s
 l : Prandtl eddy length scale, m
 L : x-direction characteristic length of eddy, m
 m, q : constants of equilibrium relation (9)
 n : exponent
 N : agitation speed, s⁻¹
 Re : Reynolds number, $N d^2 / \nu$
 s : surface renewal rate, s⁻¹
 Sc : Schmidt number, ν / D
 Sh : Sherwood number, $k d / D$
 t : time, s
 u : x-component velocity of eddy very close to the interface, m/s
 v : y-component velocity of eddy, m/s
 V_m : maximum amplitude of u, m/s
 W : mass fraction composition, wt%
 x : distance along the interface, m
 y : depth from the interface, m

Greek Letters

α, β : defined by eqn (10)
 ϵ : rate of energy dissipation, m²/s³
 μ : viscosity, Poise
 ν : kinematic viscosity, m²/s
 ρ : density, Kg/m³
 σ : interfacial tension, N/m

Subscripts

b : bulk
 eq : equivalent
 i : phase i
 m : maximum value
 o : organic phase

s : surface
 w : water phase

Superscripts

0 : initial value
 * : equilibrium

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