EXPERIMENTAL STUDIES ON MASS TRANSFER IN CONTINUOUS AND DISPERSED PHASES FOR SINGLE DROPS MOVING THROUGH LIQUID FIELD AT INTERMEDIATE REYNOLDS NUMBERS

MANABU YAMAGUCHI AND TAKASHI KATAYAMA
*Department of Chemical Engineering, Faculty of Engineering
Science, Osaka University, Toyonaka 560*

Rates of the mass transfer in both continuous and dispersed phases were measured on single non-oscillating liquid drops moving through quiescent liquid field at Reynolds numbers ranging from 40 to 1200. The mass transfer experiments of both phases were performed on systems having high interfacial tension and iodine was used as the transferred material. For the mass transfer rate in the continuous phase, the experimental data are compared with analytical solutions obtained by use of the interfacial velocity of a drop proposed by Winnikow and Chao. For the mass transfer rate in the dispersed phase, the measured results are examined in the light of some theories proposed by other investigators. In both cases, the agreements between the experimental and the theoretical values are not satisfactory. Empirical equations for mass transfer coefficients in continuous and dispersed phases are obtained for the region of the intermediate Reynolds numbers.

The drag coefficients of the drops were also measured. Although the experimental values are higher than predicted by Winnikow and Chao, the data show good agreement with the values from the literature.

Introduction

Many investigators have studied experimentally and theoretically mass transfer rates between single drops and its liquid surroundings. A large number of the studies have been performed on the systems in low Reynolds number regions^{3,9,14,23-27,33,40,46}.

For the cases when drops are moving with oscillation through quiescent liquid fields, we presented the experimental results on the mass transfer rates in previous papers^{44,46}. The results showed satisfactory agreement with the theoretical analysis of Angelo *et al.*¹⁾

For the mass transfer rates at the intermediate Reynolds number region*, theoretical studies are very difficult because of mathematical complication of the governing equations. Therefore, the theoretical analysis for the mass transfer rate in the continuous phase have been developed by various approximate analytical methods^{5,14,26,28,33,34,38,42}. For the case of the mass transfer rate in the dispersed phase, there is an analysis obtained by Harada and Eguchi¹⁷⁾. Experimental

studies for the mass transfer rates in the continuous and dispersed phases are also difficult in the region because the moving behavior of drops is effected sensitively by trace impurities and slight disturbance of liquid field, etc. There are few reliable data taken for the systems of high interfacial tension.

The present paper is composed of three sections. The Section 1.1 deals with the drag coefficients of single drops moving with internal circulation through quiescent liquid fields in the region of intermediate Reynolds number. In Sections 1.2 and 1.3, the mass transfer rates in the continuous and dispersed phases for the drops in the region of present interest are discussed, respectively. The measurements of the mass transfer rates were performed for the systems of high interfacial tension, and iodine was used as the solute which was transferred from an aqueous glycerin solution to an organic solvent phase.

1. Experimental

The distribution coefficient of iodine between an aqueous liquid phase and an organic solvent phase was large in favor of the organic solvent phase as reported in a previous paper⁴⁴⁾ that the resistance to the mass transfer between the phases could be considered to be exclusively on the aqueous liquid phase. Each of the organic chemicals except liquid paraffin

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* The region of intermediate Reynolds number used in this work is defined as the region in which Reynolds number is more than 10 and less than the critical Reynolds number of drop oscillation.

Table 1 Liquid-liquid systems used in the experiment

	Dispersed phase	Continuous phase
System 1	Benzene + 55.7wt% CCl ₄	Aqueous sol. of 40.5wt% glycerin
System 2	Liq. paraffin + 28.3wt% Benzene + 44.0wt% CCl ₄	Aqueous sol. of 29.6wt% glycerin
System 3	Aqueous sol. of 12.3wt% glycerin	Benzene + 25.0wt% CCl ₄
System 4	Distilled water	Benzene + 21.9wt% CCl ₄
System 5	Distilled water	Benzene + 25.0wt% CCl ₄ + 30.8wt% Liq. paraffin

Table 2 Physical properties of the liquid-liquid systems used

System	Temp. [°C]	Density [g/cc]	Density ratio Y [—]	Viscosity [g/cm·sec] × 10 ²	Viscosity ratio X [—]	Parameter b [—]	Interfacial tension [dyne/cm]	Diffusion coefficient* [cm ² /sec] × 10 ⁸
		Disp. phase	Cont. phase	Disp. phase	Cont. phase			
System 1	20	1.148	1.094	0.769	3.693	0.208	1.79	45.4
System 2	25	1.081	1.070	1.834	2.090	0.877	2.38	40.0
System 3	20	1.028	0.989	1.383	0.706	1.960	3.25	33.5
System 4	20	0.998	0.974	1.031	0.711	1.450	2.86	38.8
System 5	20	0.998	0.985	1.031	1.652	0.624	2.16	39.5

* The value estimated by the Wilke-Chang correlation⁴¹⁾

and glycerin was purified by a rectification column. Laboratory deionized water was distilled in an all-glass flask. The experimental apparatus and procedure used in both experiments were essentially the same as described in the previous paper⁴⁴⁾, but a drop collector used for the experiment in 1.2 was smaller than that in the previous one. The device applied for the formation of drops of a desired size was a glass capillary with a suitable diameter, which was made for every run. The frequency of the drop formation was every 5–8 seconds for a drop so as to eliminate the effect of disturbance of continuous liquid field produced by a proceeding drop on the succeeding one. The effective traveling distances of non-oscillating drops in a stationary liquid field were 3, 30 and 50 cm for the experiment in 1.2, and 5 cm and 10–40 cm at 10 cm intervals in 1.3.

Summaries of the experimental systems and their physical properties measured at each experimental temperature are given in **Tables 1** and **2**, respectively.

1.1 Moving behavior of non-oscillating drops

It is convenient to represent the experimental results as a dimensionless plot of drag coefficient versus Reynolds number not only to compare them with the behavior of solid spheres, but also to examine the influence of fluid properties on the moving behavior of the drops.

For the cases when circulation currents within a drop are caused by the friction drag of the outer fluid and the Reynolds number of the drop is high, Winnikow and Chao⁴³⁾ derived theoretically the viscous and pressure drag coefficients and obtained the total drag coefficient as the following functional form,

$$C_D = f(Re, b, \theta_s) \quad (1)$$

where b is a dimensionless parameter proposed by Chao⁴⁾ and θ_s is the polar angle at the flow separation measured from the frontal stagnation point on the

Key	b	liquid-liquid system used	investigator
▲	2.27	Chlorobenzene drop in water	Winnikow <i>et al.</i> ⁴³⁾
●	2.38	Bromobenzene drop in water	"
○	3.45	<i>m</i> -Nitrotoluene drop in water	"
▼	2.19	Toluene + 24.5 wt% CCl ₄ drop in water	Hayakawa <i>et al.</i> ¹⁹⁾
◆	2.22	Toluene drop in water	"
◇	2.25	Toluene drop in water	Harada <i>et al.</i> ¹⁷⁾
△	2.25	Benzene drop in water	Fujinawa <i>et al.</i> ⁸⁾
△	1.79	System 1	This work
⊙	2.38	System 2	"
○	3.25	System 3	"
▽	2.86	System 4	"
◇	2.16	System 5	"

Fig. 1 Drag coefficients of non-oscillating drops moving through liquid fields

drop surface. The θ_s -value can be estimated by setting $U_i=0$ in the following equation which was derived by Winnikow and Chao.

$$U_i = \frac{3}{2} \sin \theta \left[1 - \frac{4b}{\sqrt{3\pi Re}} \frac{\{(2/3) - \cos \theta + (1/3) \cos^3 \theta\}^{1/2}}{\sin^2 \theta} \right] \quad (2)$$

The results of the drag coefficient, C_D , measured for the liquid-liquid systems listed in Table 1 are shown in **Fig. 1**. In the figure some of the experimental data by Winnikow and Chao are also given for comparison with the data of present work. The C_D -

Fig. 2 Comparisons of mass transfer rates in the continuous phase between the experimental values and the values calculated by Eqs. (5) and (6) for the System 1

Fig. 3 Comparisons of mass transfer rates in the continuous phase between the experimental values and the values calculated by Eqs. (5) and (6) for the System 2

values for the System 2 show good agreement with their values for bromobenzene drops in water as shown in Fig. 1. In this case the b -value of the System 2 was identical with that used by them. On the other hand, their experimental values for the system of m -nitrotoluene drops in water are somewhat lower than our data of the System 3 though the b -value of their system was higher than that of the System 3. The C_D -values given by the relation of Eq. (1), which are shown by solid lines in Fig. 1, are considerably lower than the experimental values. The reason for the discrepancy is not clear, but it may be considered that the theoretical analysis of Chao⁴⁾ has not taken account of the effect of interfacial tension, and drag force over the rear portion behind the flow separation on the drop surface is neglected in the relation of Eq. (1).

1.2 Mass transfer rate in continuous phase

Theoretical analysis on motion and mass transfer for single non-oscillating drops is difficult at $Re \gg 1$ because of the mathematical complication of the

governing equations. The equation for the mass transfer rate in the continuous phase around the drops in the region has been developed by various approximate analytical methods^{5,14,26,28,33,34,38,42)}. Experimental studies for the mass transfer rate have been reported by many investigators and some empirical correlations have been obtained^{9,12,16,37)}. However, most of the mass transfer data were obtained for partially miscible binary systems having low interfacial tension^{9,14,21,23)} and few reliable experimental data were taken for systems of high interfacial tension.

In this section, experimental studies are shown on the mass transfer rate in the continuous phase for single non-oscillating drops at Reynolds numbers ranging from 40 to 300. The Systems 1 and 2 shown in Table 1 were used in the experiment, and iodine used as the solute was transferred from an aqueous continuous phase to an organic solvent drop phase.

Experimental results and discussion: Mass transfer coefficient, k_c , in the continuous phase around a drop during the steady motion period was calculated by,

$$k_c = N / \{A(C_c - C^*)t\} \quad (3)$$

where t is the residence time and A is the surface area of the drop assumed an equivalent spherical diameter. The value of N in Eq. (3) was evaluated as the difference between the total iodine quantity transferred into a drop for a given falling distance and that for the falling distance of 3 cm so as to eliminate the end effects in this experiment. The driving force, $(C_c - C^*)$, was almost unchanged (less than 1%) during each of the experimental runs because liquid volume of the continuous phase was very large in comparison with the one of the dispersed phase, and the solute quantity transferred was very small. The experimental results of the mass transfer rate for the Systems 1 and 2 are shown in Figs. 2 and 3, respectively.

At Péclet numbers greater than about 100, forced-convective mass transfer equation in the bulk phase around single non-oscillating drop moving with rapid interfacial velocity was proposed by Lochiel and Calderbank²⁸⁾ as the following general expression,

$$\overline{Sh}_c = \left(\frac{2}{\pi}\right)^{1/2} \left(\int_0^\pi U_i \sin^2 \theta d\theta\right)^{1/2} Pe_c^{1/2} \quad (4)$$

where U_i is the dimensionless interfacial velocity of the drop. In the range of Reynolds numbers of present interest, the flow separation of boundary layer on the drop surface occurs at a rear part of the drop. The location of the separation, θ_s , is calculated by setting $U_i = 0$ in Eq. (2). If the mass transfer in the outer wake region ($\theta_s - \pi$) is assumed to be zero and Eq. (2) is substituted into Eq. (4), we have the following expression for the continuous-phase mass transfer rate based on the total surface area of the drop.

$$\begin{aligned} \overline{Sh}_e = & \frac{1}{\sqrt{\pi}}(1 - \cos \theta_s) \left[\frac{1}{2} - \frac{3}{4} \cos \theta_s + \frac{1}{4} \cos^3 \theta_s \right. \\ & - \frac{36}{5(3\pi Re)^{1/2}} b \left\{ 1 - \frac{\sqrt{3}}{8} (3 - \cos \theta_s) \right. \\ & \left. \left. \times (2 + \cos \theta_s)^{3/2} \right\} \right]^{1/2} \times Pe_e^{1/2} \quad (5) \end{aligned}$$

Provided that the flow separation of boundary layer on the drop surface does not take place, the closed solution ($\theta_s = \pi$) is consistent with the solution of Lochiel²⁷) and that of Winnikow⁴²). Figures 2 and 3 represent the numerical values calculated by the closed solution (the broken lines) and the experimental values of present work. The data are considerably lower than the broken line. Even the numerical values calculated by Eq. (5) (the chain lines) are still somewhat higher than the experimental ones. As shown in these figures, the experimental values of mass transfer rate decrease considerably with decreasing Reynolds number and show the tendency of approaching to the line of Ranz and Marshall³¹). Whether this is due to minute traces of impurities or to intrinsic properties of the liquid-liquid systems used is not clear at present. Generally speaking, the presence of impurities in liquid-liquid systems results in reducing the interfacial velocity on a drop^{6,43}) and will increase solid-like behavior of the drop with decreasing drop size. According to Garner and Skelland^{10,11}), it is possible that the fluid inside drops circulates less readily with increasing the interfacial tension. In all events, it is clear that actual interfacial velocity on the drop differs from the theoretical one predicted by Eq. (2).

The mass transfer data obtained in this work and those from the literature³⁷) were well correlated by the following equation,

$$\left. \begin{aligned} \overline{Sh}_e &= 2 + f_1 Pe_e^{0.5} \\ f_1 &= \frac{1}{3(1+X)} + \frac{2+3X}{3(1+X)} \frac{Re}{(16+16X+Re)} \\ &\quad - \frac{4}{3} b \sqrt{\frac{2}{\pi}} \frac{1}{\sqrt{Re}} \end{aligned} \right\} \quad (6)$$

The functional form of f_1 in Eq. (6) was determined from a modification of the interfacial velocity on the equatorial plane of a drop proposed by Shiotsuka and Hirata³⁴), but a theoretical basis for the modification does not exist. **Figure 4** shows the comparisons of the values calculated by Eq. (6) with experimental values obtained in this work and from the literature^{32,37}). The experimental data of Redfield and Houghton³²) for bubbles were also correlated satisfactorily with the equation as shown in the figure. The differences between the experimental and the calculated values are approximately within $\pm 20\%$. When the value of viscosity ratio becomes larger, it is well known that the index of Schmidt number changes gradually from 1/2 to 1/3 because the moving be-

Fig. 4 Comparisons of mass transfer rates in the continuous phase predicted by Eq. (6) with the experimental values and those from the literature^{32,37})

havior of a liquid drop approaches that of a rigid sphere. Although the upper limitation of the viscosity ratio cannot be clarified for lack of experimental data, Eq. (6) may be applied in the range of $X < 3$ and $Re > 40$.

1.3 Mass transfer rate in dispersed phase

Mass transfer in a liquid drop is greatly influenced by the flow behavior within the drop. The mass transfer rate has been theoretically solved^{15,25,29}) only under three idealized flow patterns within the drop. The applications of the models were examined experimentally by several investigators^{13,17,24,30,35}), but the results were not satisfactory in the region of intermediate Reynolds numbers. Although some modifications for the models have been attempted^{17,24,30,39}), they are not always useful because they need experimental parameters.

In this experiment, the mass transfer rates within the drops moving through quiescent liquid fields in a region of intermediate Reynolds numbers were measured by use of the Systems 3, 4 and 5 shown in Table 1. The iodine used as the solute was transferred from an aqueous drop phase to a continuous solvent phase.

Experimental results and discussion: Mass transfer coefficient, k_d , within a drop during the period of steady movement is given from a differential material balance and the boundary conditions as follows,

$$k_d = -\frac{V}{A} \frac{d}{dt} \ln \left\{ \frac{(C_d - C^*)}{(C_d^i - C^*)} \right\} \quad (7)$$

An example of the experimental results for the System 3 is shown in **Fig. 5**, in which the values of $-\ln \{ (C_d - C^*) / (C_d^i - C^*) \}$ are plotted against the Fourier number. The solid line in the figure is the one fitted to the experimental values by use of the method of least squares. The broken line calculated by use of Kronig

Fig. 5 Relation between fractional unextraction in a drop and Fourier number

Fig. 6 Comparisons of mass transfer rates in the dispersed phase between the values calculated by Eq. (5) and the experimental values of the System 3 and those from the literature^{8,17,20}

Fig. 7 Correlation between mass transfer rate in the dispersed phase for non-oscillating drop and Fourier number

and Brink's model²⁵) differs greatly from the solid line as shown in the figure. The results obtained in this work were represented by similar solid lines for the whole series runs. A local value of k_d at any given dimensionless time was determined from the derivative of the curve.

Taunton and Lightfoot³⁶) obtained an equation for the mass transfer rate in the dispersed phase at high Reynolds number by using the generalized surface

stretch modification of the penetration theory. In the derivation of the equation, they used the corrected velocity profile developed by Chao⁴) as the interfacial velocity of the drop. On the case of a large contact time, they obtained an equation for the Sherwood number in the dispersed phase averaging between the frontal stagnation point and the boundary layer separation of the drop. Their equation is identical with Eq. (5) when it is rearranged. In Fig. 6, the results calculated by Eq. (5) (the solid lines) are compared with the experimental values of the System 3 and those from the literature^{7,8,17,18,19,20}). The equation given by Higbie²²) (the broken line) and that by Kronig and Brink²⁵) (the chain line) are also shown in the figure for comparison. All k_d -values of the Sherwood number in the figure were obtained for the lapse of 2.5 sec after detaching of drops from the nozzle. The data obtained in this work and those of Harada and Eguchi^{7,17,18}) are considerably lower than the solid lines as shown in Fig. 6. These data, however, approach to the line of Higbie with increasing Reynolds number. The experimental values obtained by other investigators^{8,19,20}) are, in contrast with our data, higher than the solid line calculated by Eq. (5).

As theoretical analysis for the mass transfer rate in the region of present interest is difficult, an empirical correlation of the results obtained in this work was attempted by dimensional analysis. Figure 7 shows the relation between the experimental dimensionless group, $\bar{Sh}/(Ca \cdot X^{0.5})$, for the mass transfer rate and the experimental Fourier number, Fo . As the results, an empirical correlation for the mass transfer rate in dispersed phase was obtained as follows,

$$\bar{Sh}_d = 3.0 \times 10^3 Fo^{-0.5} X^{0.5} Ca \quad (8)$$

The solid line in the figure indicates the correlation expressed by Eq. (8). The differences between the correlation and the experimental results were approximately within $\pm 30\%$ as shown in Fig. 7. The broken line in the figure is an empirical correlation obtained by Hayakawa and Shigeta²⁰), which gives a higher value than that of Eq. (8). The reason for the discrepancy is not clear, but it may be deduced that their drops might probably rise with abnormal motions such as zig-zag or wobbling since the C_D -values of their original data¹⁹) were higher than those of solid sphere as shown in Fig. 1.

Conclusion

By use of the liquid-liquid systems having high interfacial tension, experimental studies of mass transfer rates in the continuous and dispersed phases were performed in a range of intermediate Reynolds numbers. The conclusions of the present work are summarized as follows.

1) The drag coefficients measured for drops of organic liquids falling through aqueous glycerin solutions were in good agreement with experimental data by Winnikow and Chao, but those for aqueous drops falling through organic solvent phases showed somewhat higher values than their data. The obtained data gave considerably higher values than the ones calculated by the theoretical equation of Winnikow and Chao.

2) The experimental data in the continuous phase for non-oscillating drops were expressed satisfactorily by an empirical equation, Eq. (6). The equation also correlates the data for high interfacial tension systems available from the literature in the range of $X < 3$ and $Re > 40$.

3) The experimental mass transfer rates in the dispersed phase showed the time dependency as reported in the literature. The Sherwood numbers obtained experimentally in the range of $0.62 < X < 1.97$ and $50 < Re < 1200$ have been correlated in terms of Fourier number, Capillary number and viscosity ratio as Eq. (8).

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Nomenclature

A	= interfacial area of a drop	[cm ²]
b	= $(2+3X)/(1+\sqrt{X \cdot Y})$	[—]
C	= iodine concentration in aqueous phase	[g-mol/cc]
Ca	= Capillary number, $\mu_c U_i / \sigma_i$	[—]
C_D	= drag coefficient, $4 \rho_d - \rho_c g \cdot d_e / 3\rho_c U_i^2$	[—]
$\mathcal{D}$	= diffusivity of iodine	[cm ² /sec]
d_e	= equivalent spherical diameter of a drop	[cm]
Fo	= Fourier number, $4\mathcal{D}t/d_e^2$	[—]
k	= mass transfer coefficient	[cm/sec]
N	= amount of transferred solute	[g-mol]
$Pé$	= Péclet number, $d_e U_i / \mathcal{D}$	[—]
Re	= Reynolds number, $\rho_c U_i d_e / \mu_c$	[—]
Sh	= Sherwood number, $kd_e / \mathcal{D}$	[—]
t	= residence time of a drop moving through liquid field	[sec]
U_i	= dimensionless interfacial velocity	[—]
U_t	= terminal velocity	[cm/sec]
V	= drop volume	[cc]
X	= viscosity ratio, μ_d / μ_c	[—]
Y	= density ratio, ρ_d / ρ_c	[—]
θ	= polar angle measured from frontal stagnation point	[radian]
μ	= viscosity	[g/cm·sec]
ρ	= density	[g/cc]
σ_i	= interfacial tension	[dyne/cm]

<Subscripts>

c	= continuous phase
d	= dispersed phase
s	= separation point

<Superscripts>

—	= average
i	= initial condition
*	= equilibrium value

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MASS TRANSFER TO A ROTATING SPHERE IN A STREAM

TAKESHI FURUTA, MORIO OKAZAKI AND RYOZO TOEI
Department of Chemical Engineering, Kyoto University, Kyoto 606

The local and average mass transfer coefficients to a rotating sphere were measured by electrochemical method, when the angle α between the axis of rotation and the direction of the uniform stream were both 45° and 90° . For measurements of the local mass transfer rates, circular isolated electrodes of 2 mm dia. were used, and the time averaged values and the variations of the local mass transfer rates were obtained by processing the signals from each isolated electrode during some revolutions. Moreover, on the basis of the measurements of average mass transfer coefficients, new empirical equations were obtained with which they have been correlated well for $\alpha=0^\circ$, 45° and 90° .

Introduction

The study on heat and mass transfer to a rotating sphere in a stream is important in the analysis of heat or mass transfer between the circumferential fluid and the particles suspended in it³⁾. Several studies have been reported on heat or mass transfer to/from a rotating sphere placed in a uniform stream. From a hydrodynamic point of view, these studies should be divided into two classes. When the angle α between the uniform stream and the axis of rotation of the sphere is equal to zero, the velocity profile around the sphere shows axially symmetry. On the other hand, when $\alpha \neq 0^\circ$, the velocity profile is completely three dimensional so that it becomes very difficult to study it theoretically. In the case of $\alpha=0^\circ$, Tanaka and Tago⁵⁾ suggested a semi-empirical equation for the average mass transfer coefficients, which could correlate the experimental results well. Also the present authors²⁾ obtained the local mass transfer rates by solving the laminar boundary layer equations, and the numerical results showed them in good agreement with the measurements. On the other hand, for $\alpha=$

90° , Noordsij and Rotte⁴⁾, and Tanaka and Tago⁵⁾ measured the average Sherwood numbers, and moreover Eastop¹⁾ obtained the average Nusselt numbers. However all these investigations have been no more than the measurements of average heat or mass transfer coefficients.

In the present work, the local mass transfer rates of a sphere rotating in a uniform stream were measured for $\alpha=90^\circ$ and 45° by means of the so-called electrochemical method. At the same time, also the average mass transfer coefficients were obtained to provide some discussions about the effects of α .

1. Experimental Apparatus and Procedure

The experimental apparatus is diagrammatically shown in **Fig. 1(a)**. Almost all the parts except for the test section are the same as that in our previous study²⁾. The test section used for $\alpha=45^\circ$ is illustrated in **Fig. 1(b)**.

In the previous investigation²⁾, the local mass transfer rates to an isolated electrode were stationary during a revolution of the sphere, since the velocity profile around the sphere was symmetric about the axis of rotation. However, because α is not zero in the present work, the velocity profile becomes three

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