

EFFECT OF INERT PARTICLE CONCENTRATION ON MASS TRANSFER BETWEEN PARTICLES AND LIQUID IN SOLID-LIQUID TWO-PHASE UPFLOW THROUGH VERTICAL TUBES AND IN STIRRED TANKS

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Key Words: Mass Transfer, Ion Exchange, Agitation, Two Phase Flow, Particle, Energy Dissipation, Turbulence

Mass transfer coefficients between particles and liquid in the region of high particle concentration were obtained by the use of both active and inert particles suspended in solid-liquid two-phase upflow through vertical tubes and in stirred tanks. When adding inert particles of low density such as polystyrene and anion-exchange resin beads, mass transfer coefficients decreased with increasing particle concentration, remarkably above concentrations from 20 to 30%. Meanwhile, the coefficients increased in two-phase tube flow and decreased in stirred tanks when using glass beads. The experimental results were correlated with parameters including (1) the kinematic viscosity of slurry and (2) the effective energy dissipation rate, based on the concept that only a part of the energy supplied contributes to the mass transfer. The first method was successfully applied only to the system containing low-density particles, and the second to systems having either high- or low-density particles. The effectiveness of energy dissipation on mass transfer at high particle concentration is discussed in some detail.

Introduction

Mass transfer between particles and liquid at high particle concentration is employed in many practical chemical processes. However, most experimental researches on mass transfer have been restricted to the region of low particle concentration where the interaction among particles is negligibly small. For example, a particle concentration less than 1% was employed in a series of our work^{11,16,19,20)} on solid-liquid two-phase flow through vertical and horizontal tubes, and less than 5% in the previous work^{4,6)} for horizontal tubes. In these studies, no effect was observed on mass transfer between particles and liquid in such a low range of particle concentration. The same low order of particle concentration was adopted in the previous work^{2,3,13,14,22)} for stirred tanks, where no effect of particle concentration was also reported. On the other hand, some investigators insisted that a considerable effect of particle addition was observed even in the low concentration range on mass transfer between particles and liquid. Recently, Boon-Long *et al.*¹⁾ revealed that mass transfer coefficients drastically decreased in the dissolution of

benzoic-acid particles of 3.02 mm diameter in stirred tanks with an increase in particle concentration from 1.5 to 3%. They claimed that it would result from the incomplete dispersion of particles or the damping of turbulence level due to the presence of particles in the vicinity of an impeller. Yagi *et al.*²⁴⁾ also found a decrease in the mass transfer coefficient to the extent of about one-tenth for dissolution of calcium hydroxide particles of 25.5 μ m diameter in the particle concentration range of 0.2 to 2%. They explained the experimental results by a model which assumed the particles to be captured in turbulent eddies.

As mentioned above, a difference is seen among the previous results concerning the effect of particle concentration, even at low level, on mass transfer. As for high particle concentration, little work has been done.

The purpose of this work is to investigate the effect of particle concentration on mass transfer between particles and liquid in solid-liquid two-phase upflow through vertical tubes and in stirred tanks, where their macroscopic flow patterns seem to be considerably different from each other, by use of the following particle systems: active particles (beads) of cation-exchange resin at a concentration of less than 1%; inert particles of polystyrene, anion-exchange resin, and glass beads at concentrations of 0–50%.

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1. Experimental Apparatus and Procedure

Figure 1 shows the experimental apparatus for two-phase upflow through vertical tubes. The details are the same as in the previous work^{16,19} except for the injection system of inert particles, which is constructed so as to supply the inert particles at a certain rate. The holder of inert particles (5) is made of polyvinyl chloride resin pipe of 0.25 m in diameter and 2.3 m in length. The reactor (1) is made of Plexiglass, 3.00 cm in inner diameter and 5.1 m in reaction length. Experimental procedure is nearly the same as that in the previous work.¹⁶ Active and inert particles are simultaneously supplied into the tube, forming two-phase flow with the electrolyte solution. The total amounts of solution and particles were separately sampled at the reactor exit without recycling. Slurry velocity was determined from the sampled weight. The concentrations, i.e., the volume fractions, of both active and inert particles in the outlet solution were calculated from the weight of each particle separated by density difference. Temperature range is from 12 to 23°C.

Figure 2 shows a schematic sketch of stirred tanks, sizes of which are listed in Table 1. The stirred tanks, made of glass or Plexiglass, are designated A, B, and C, and have different capacities. The impeller is a 6-flat blade turbine. Impeller speed was measured by a stroboscope. The impeller's power requirement was measured by a torsion-spring torque meter manufactured by Yamasaki Ltd. A run started with charging a given amount of inert particles and potassium chloride solution, the concentration of which was 5×10^{-4} kmol/m³,¹⁶ into the tank. The impeller speed was set so as to maintain complete suspension. Next, active particles were put into the tank, and the solution was periodically sampled with a pipet, the top of which was covered with Saran cloth to filter the particles. Ion concentrations of the sampled solution were determined by atomic absorption analysis. Experimental temperature was 20 or 30°C.

The active particle used is of styrene-type sulfonic acid cation-exchange resin and of sodium form throughout the experiments. The properties of both active and inert particles are shown in Table 2. The diffusivity for Na⁺-K⁺ exchange was estimated with Kataoka's equation, considering the effect of electric field.⁹⁾

2. Calculation of Mass Transfer Coefficient

The stoichiometry for cation exchange is expressed by the following equation:



Taking the mass balance for the ion species B on the assumption that the liquid-side diffusion is the

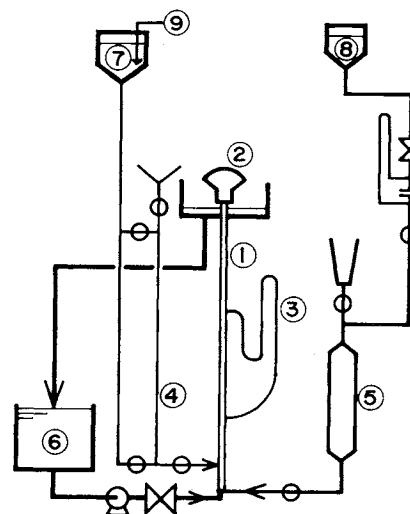


Fig. 1. Schematic diagram of experimental apparatus for two-phase tube flow. (1), reactor tube; (2), particle separator; (3), manometer; (4), active particle holder; (5), inert particle holder; (6), stock tank; (7), (8), head tank; (9), carbon dioxide-free air.

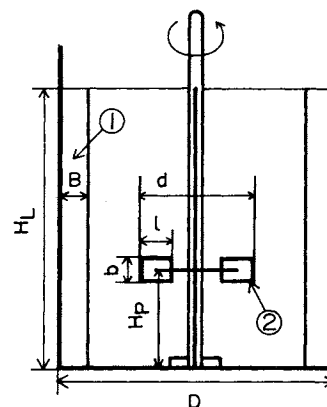


Fig. 2. Schematic representation of stirred tank. (1), baffle plate; (2), impeller.

Table 1. Dimensions of tanks and impellers (refer to Fig. 2)

		A	B	C
Tank	Slurry volume [l]	0.7	1.7	5.4
	$D=H_L$ [mm]	96	130	190
	B/D [—]	1/10	1/10	1/10
Impeller	d [mm]	28	55	55
	b [mm]	5	10	10
	l [mm]	8	14	14
	d/D [—]	0.29	0.42	0.29
	H_p/H_L [—]	1/3	1/3	1/3

rate-determining step, the liquid-side mass transfer coefficient in two-phase tube-flow is formulated by Eq. (2).

$$\bar{k}_L = \bar{U}_p (1/L_T) (D_p/6) (V_w/V_p) \ln(c_0/c_e) \quad (2)$$

where

Table 2. Properties of particles

	Material (Symbol)	Diameter [μm]	Density [kg/m^3]
Active	Cation-exchange resin (DIAION SK1B)	941	1280
		805	1280
		568	1280
	Glass (G)	1710	2500
Inert	Glass (G)	900	2520
		540	2490
		562	1050
	Polystyrene (PS)	325	1050
		290	1050
	Anion-exchange resin (IR) (DIAION SA20AP)	566	1150

$$\bar{k}_L = \frac{\int_s N k_L ds}{\int_s N ds} \quad (3)$$

$$\bar{U}_p = \frac{\int_s N U_p ds}{\int_s N ds} \quad (4)$$

The average velocity of active particles \bar{U}_p is estimated by our empirical equation,¹⁷⁾ which is ascertained beforehand to apply even at a high concentration of particles.

The mass transfer coefficient for stirred tanks is expressed by the following equation:

$$\bar{k}_L = -\{(1 - m_{ct})D_p/6m_{ct}\theta\} \ln(c_e/c_0) \quad (5)$$

3. Experimental Results and Discussion

3.1 Mass transfer coefficient

Figures 3 and 4 illustrate some examples of experimental results for two-phase tube flow including glass and polystyrene beads as inert particles, respectively, obtained by varying the concentration of inert particles at a constant slurry velocity. Figure 3 shows the increase in mass transfer coefficient with addition of glass beads. The tendency becomes more remarkable with an increase in diameter of the inert particles. On the contrary, a decrease in mass transfer coefficient is observed with the addition of polystyrene beads as shown in Fig. 4. Similar behavior was seen for the addition of anion-exchange resin beads, though it is not shown here.

Figure 5 shows the experimental results for stirred tanks, where the concentration of inert particles was varied at specified impeller speeds. The mass transfer coefficient for stirred tanks decreases with addition of glass beads as inert particles in contrast with the result for two-phase tube flow. In the case of polystyrene beads, the mass transfer coefficient decreases remarkably with increasing particle concentration as in the case of two-phase tube-flow. The experimental results lead to the following two conclusions. Firstly,

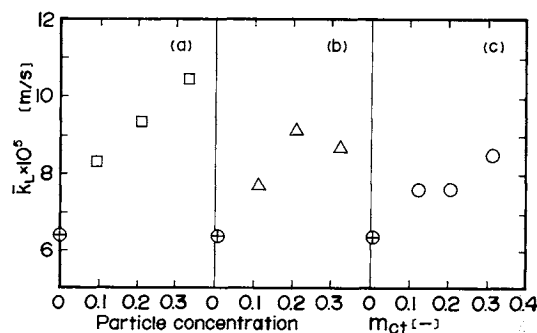


Fig. 3. Effect of particle concentration on mass transfer coefficient in two-phase tube flow with glass beads as inert particles; diameter of active particles is $568 \mu\text{m}$ and those of inert particles are $1710 \mu\text{m}$ (a), $900 \mu\text{m}$ (b), and $540 \mu\text{m}$ (c); average slurry velocity is 0.72 m/s ; \oplus denotes values for active particles only.

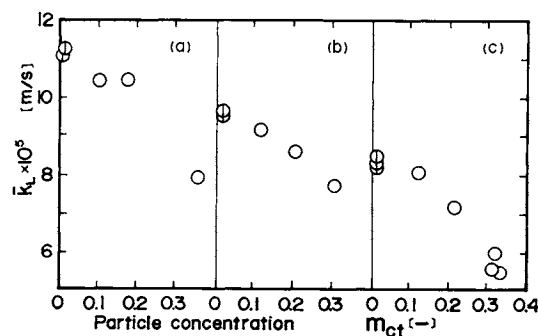


Fig. 4. Effect of particle concentration on mass transfer coefficient in two-phase tube flow with polystyrene beads as inert particles; diameters of active and inert particles are $941 \mu\text{m}$ and $562 \mu\text{m}$, respectively; average slurry velocities are 1.43 m/s (a), 1.03 m/s (b), and 0.71 m/s (c); \oplus denotes values for active particles only.

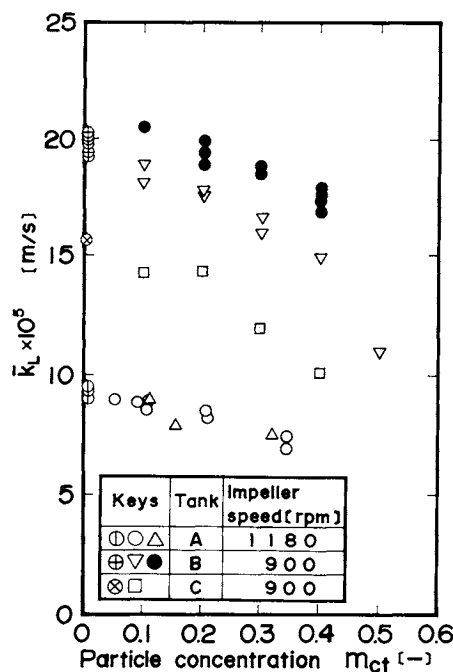


Fig. 5. Effect of inert particle concentration on mass transfer coefficient in stirred tanks: glass beads \bullet ; polystyrene \circ , \triangle , ∇ , \square ; refer to Fig. 7 for details of keys.

with the addition of such low-density particles as polystyrene and ion-exchange resin beads, the mass transfer coefficients begin to decrease remarkably in the particle concentration range of 20–30%, which is considerably higher than those of Boon-Long *et al.*¹¹⁾ and Yagi *et al.*²⁴⁾ In the present mass-transfer systems, the particles were completely dispersed in the suspension and the diameters of active particles were larger than the scale of turbulent eddies. Therefore, the decrease in mass transfer coefficient would mainly be due to the damping of turbulence level. Secondly, in the case of adding glass beads of high density, the dependence of mass transfer coefficient on particle concentration is quite different between two-phase tube flow and stirred tanks. This fact is related to the difference in flow characteristics in the two systems, which will be discussed in the next section.

3.2 Correlation

1) **Energy dissipation rate** We have recently proposed the following correlation equation of mass transfer coefficient for both vertical and horizontal two-phase tube flow¹¹⁾ in the low particle concentration region.

$$Sh = 2 + 0.47(\varepsilon^{1/3} D_p^{4/3} / \nu)^{0.63} Sc^{1/3} \quad (6)$$

Equation (6) proved to be applicable to other various mass transfer systems including stirred tanks, bubble columns, and even single particles, fixed beds and fluidized beds.^{10,18)}

Now we will apply this correlation method to the present mass transfer systems at high particle concentration. The energy dissipation rate included in the specific power group $(\varepsilon^{1/3} D_p^{4/3} / \nu)$ is calculated with the energy supplied to the turbulent field, where particles are suspended, by the following equations: for stirred tanks,

$$\varepsilon = P / V \rho_w \quad (7)$$

for solid-liquid two-phase upflow through a tube,

$$\begin{aligned} \varepsilon = & m_c U_r g(\rho_p - \rho_w) / \rho_w + m'_c U'_r g(\rho'_p - \rho_w) / \rho_w \\ & + (2f U_w^3 / D_T)(1 - m_c U_r / U_w - m'_c U'_r / U_w) / \\ & (1 - m_c - m'_c) \end{aligned} \quad (8)$$

obtained by applying the energy balance of Kada and Hanratty⁸⁾ to the present system, where nonprime and prime denote active and inert particles respectively.

Figure 6 shows the effect of particle concentration on the energy dissipation rate for stirred tanks and solid-liquid two-phase tube flow respectively, calculated by Eqs. (7) and (8), where agitation power P in Eq. (7) was measured experimentally, and relative velocities between particles and liquid U_r in Eq. (8) were estimated by the equation of Ishii and Zuber.⁷⁾ As the figure shows, the energy dissipation rate increases more remarkably for glass beads than for

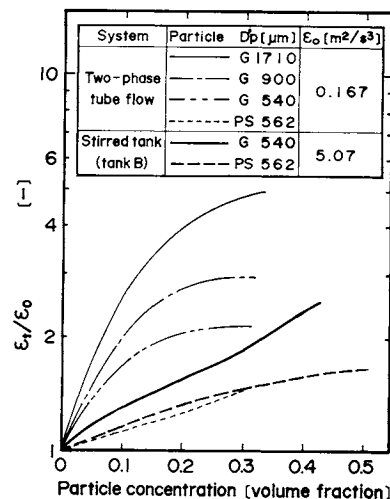


Fig. 6. Change in $\varepsilon/\varepsilon_0$ with particle concentration calculated by Eqs. (7) and (8).

polystyrene beads since the higher density causes a larger contribution to the energy dissipation.

2) **Correlation with kinematic viscosity of slurry**

Figure 7 shows the correlation of experimental results for two-phase tube flow (a) and stirred tanks (b) with specific power group $(\varepsilon^{1/3} D_p^{4/3} / \nu_m)$ including ε calculated by Eqs. (8) or (7) and kinematic viscosity of slurry ν_m estimated with the viscosity of slurry by Eq. (9)⁷⁾ ($(m_{cl})_{max} = 0.62$) and the density of slurry by Eq. (10).

$$\mu_m = \mu_w \{1 - m_{cl} / (m_{cl})_{max}\}^{-2.5(m_{cl})_{max}} \quad (9)$$

$$\rho_m = (1 - m_{cl})\rho_w + m_c \rho_p + m'_c \rho'_p \quad (10)$$

For two-phase tube flow, Fig. 7(a) indicates that the observed values with addition of glass beads disagree with those without addition of inert particles, while those with polystyrene and anion-exchange resin beads show good agreement. As for stirred tanks, Fig. 7(b) shows that all the data were satisfactorily correlated in spite of the difference in kinds of inert particles and sizes of stirred tanks. Equation (6) is included in Fig. 7 as a reference. Consequently, correlation with the kinematic viscosity ν_m is successfully achieved in the system including particles with low density such as polystyrene and anion-exchange resin beads. However, it is not always good in the system including such high-density particles as glass beads. The reason for the above correlation characteristics is explained as follows.

Mass transfer between liquid and particles in turbulent suspension should be promoted by turbulence of the liquid in general. At the same time, the liquid turbulence can be damped, making it less effective for mass transfer, by an increase in particle concentration.^{12,15,21,23)} The kinematic viscosity of slurry is considered as a parameter to express the effect of damping of turbulence on mass transfer between

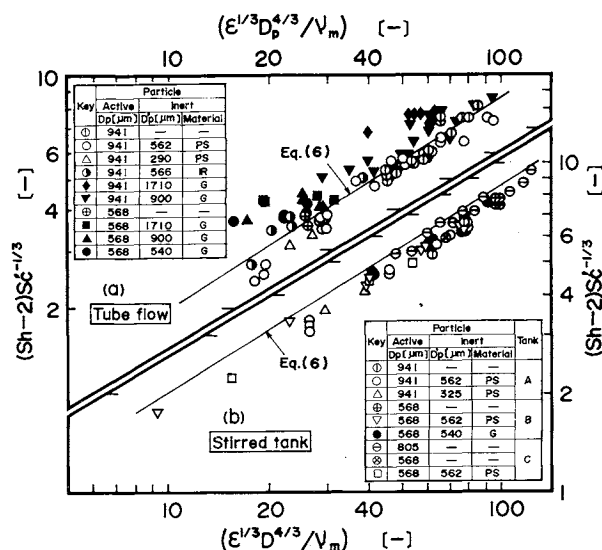


Fig. 7. Correlation with Sherwood number to specific power group and Schmidt number for two-phase tube flow (a) and stirred tanks (b) by use of kinematic viscosity of slurry: $Sc' = 406-5050$.

liquid and particles. When the diameter of particles suspended is large and its density is high, however, gravity and inertia cause a large relative velocity between particles and liquid which contributes to the compensation of damping and to an increase in turbulence.

3) Correlation with effective energy dissipation rate
A part of energy supplied into the turbulent field is dissipated into heat in the vicinity of particle surface. Therefore, it is considered that only a part of energy supplied contributes to mass transfer between particles and liquid as discussed above. Now, on the basis of the concept of the energy effective for mass transfer, effective energy dissipation rate ε_e is defined by the following equation.

$$\varepsilon_e = \varepsilon \exp(-\alpha m_{cl}) \quad (11)$$

where α is a constant and a measure of damping of turbulence, affected by such various factors as the properties of particle and liquid, the energy-supplying method, and so forth. The larger the value of α is, the smaller the energy effective for mass transfer to the energy supplied is.

Figure 8 shows the correlation of all the experimental results with specific power group including effective energy dissipation rate. The values of kinematic viscosity used are those for liquid. In Fig. 8(a), the observed values for two-phase tube flow coincide with the empirical equation Eq. (6) obtained under the conditions of low particle concentration and the presence of active particles only. It assures the availability of Eq. (6) to the high particle concentration region with the present correlation method. Figure 8(b) shows that the observed values for stirred tanks lie between the equations of Levins-Glastonbury¹⁴ and Harriott⁵ and are about 9% smaller than Eq. (6). However, taking into account of experimental error, it would be allowed to apply Eq. (6) also to the stirred tanks. The values of α in Eq. (11), determined so as to keep the least standard deviation for the correlation, are as follows: for glass beads, 0.0 for two-phase tube flow, 3.6 for stirred tanks; for anion-exchange resin beads, 5.2 for two-phase tube flow; for polystyrene beads, 6.6 for both two-phase tube flow and stirred tanks.

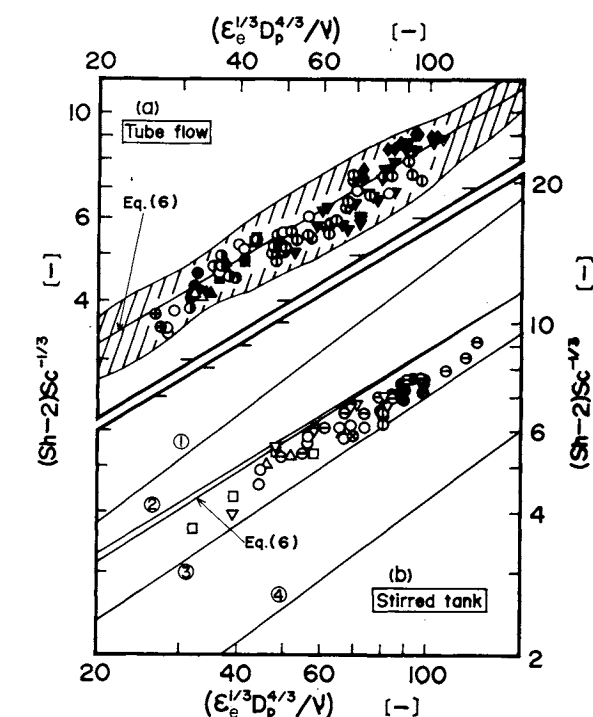


Fig. 8. Correlation with Sherwood number to specific power group and Schmidt number for two-phase tube flow (a) and stirred tanks (b) by use of effective energy dissipation rate: $Sc = 406-1030$. (Keys are same as those in Fig. 7) /// : area covered with observed data for two-phase tube flow with low concentration of active particles only.¹⁶ Lines: (1), Sano *et al.*²²; (2), Levins and Glastonbury¹⁴; (3), Harriott⁵; (4), Calderbank and Moo-Young³.

red tanks lie between the equations of Levins-Glastonbury¹⁴ and Harriott⁵ and are about 9% smaller than Eq. (6). However, taking into account of experimental error, it would be allowed to apply Eq. (6) also to the stirred tanks. The values of α in Eq. (11), determined so as to keep the least standard deviation for the correlation, are as follows: for glass beads, 0.0 for two-phase tube flow, 3.6 for stirred tanks; for anion-exchange resin beads, 5.2 for two-phase tube flow; for polystyrene beads, 6.6 for both two-phase tube flow and stirred tanks.

The values of α for glass beads are relatively small since the density of glass beads is high and the turbulence of liquid effective for the mass transfer is least damped as discussed in the former section. Especially, in the case of two-phase upflow through a vertical tube, the turbulence generated around all the suspended particles promotes mass transfer, and the influence of damp of the turbulence is not remarkable. As for stirred tanks, on the contrary, the generating source of turbulence is limited around an impeller, and both the turbulence transported by delivered flow from the impeller and that generated around the particles are damped within the distance not so far from the impeller.^{12,21,23} That would be one of the reasons why the value of α for glass beads suspended in the

stirred tanks is larger than that in two-phase tube flow. Meanwhile, in the case of particles with low density such as anion-exchange resin and polystyrene beads, the effect of the generation of turbulence is negligibly small in comparison with that of damp of turbulence, therefore, the values of α are larger than those for glass beads. It is interesting that the values of α for polystyrene beads are the same between stirred tanks and two-phase tube flow in spite of the considerable difference in the macroscopic flow pattern. One of the reasons would exist in such a low density that it enables the particles to follow the fluid flow readily and the slurry to be microscopically homogeneous.

Concluding Remarks

Mass transfer coefficients between particles and liquid at high particle concentration were obtained by use of both active and inert particles suspended in solid-liquid two-phase upflow through vertical tubes and in the stirred tanks.

When adding inert particles of low density such as polystyrene and anion-exchange resin beads, mass transfer coefficients decreased with increasing particle concentration, remarkably above concentrations from 20 to 30%. Meanwhile, the coefficients increased in two-phase tube flow and decreased in stirred tanks when using glass beads. The experimental results were firstly correlated with the kinematic viscosity of slurry and with the energy dissipation rate calculated from the energy supplied into the turbulent field. This correlation method was successfully applied to the system containing polystyrene and anion-exchange resin beads, but not always to the glass-beads system. Secondly, the experimental results were correlated by use of the concept of effective energy dissipation rate, which proved to apply to the systems with either low- or high-density particles. All the observed values agreed with the correlation equation obtained for low particle concentration in two-phase tube flow with due consideration of specific power group. The effectiveness of energy dissipation on mass transfer at high particle concentration was discussed in terms of particle diameter, density, and method of energy-supply.

Acknowledgment

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Nomenclature

A	= ion species A	[—]
B	= ion species B	[—]
c_e	= concentration of counter-ion B at reactor exit, or at time θ	[kmol/m ³]

c_0	= concentration of counter-ion B at reactor inlet, or at time 0	[kmol/m ³]
\mathcal{D}	= diffusivity	[m ² /s]
D_p	= diameter of active particles	[m]
D'_p	= diameter of inert particles	[m]
D_T	= inner diameter of tube	[m]
f	= friction factor for smooth tube (= $0.082 Re_T^{-0.25}$)	[—]
g	= acceleration of gravity	[m/s ²]
k_L	= local liquid-side mass transfer coefficient	[m/s]
\bar{k}_L	= average liquid-side mass transfer coefficient	[m/s]
L_T	= length of tube	[m]
m_c	= concentration of active particles in tube	[—]
m'_c	= concentration of inert particles in tube	[—]
m_{ct}	= total concentration of particles in reactor tube (= $m_c + m'_c$)	[—]
$(m_{ct})_{\max}$	= total concentration of particles for maximum packing	[—]
N	= number of particles per unit reactor volume	[m ⁻³]
n_i	= impeller speed	[s ⁻¹]
P	= power input to impeller	[W]
Re_T	= Reynolds number (= $U_w D_T / \nu$)	[—]
Sc	= Schmidt number (= ν / \mathcal{D})	[—]
Sc'	= Schmidt number (= ν_m / \mathcal{D})	[—]
Sh	= Sherwood number (= $\bar{k}_L D_p / \mathcal{D}$)	[—]
U_r	= relative velocity between active particles and liquid	[m/s]
U'_r	= relative velocity between inert particles and liquid	[m/s]
U_w	= liquid velocity	[m/s]
V	= liquid volume in stirred tank	[m ³]
V_w	= sampled volume of liquid for arbitrary time interval	[m ³]
V_p	= sampled volume of active particle for arbitrary time interval	[m ³]
Z_A, Z_B	= ionic valence	[—]
α	= constant	[—]
ε	= energy dissipation rate per unit mass of liquid, defined by the energy supplied to the turbulent flow field	[m ² /s ³]
ε_e	= effective energy dissipation rate per unit mass of liquid, defined by Eq. (11)	[m ² /s ³]
ε_0	= energy dissipation rate per unit mass of liquid in the absence of particle	[m ² /s ³]
θ	= time	[s]
μ_m	= viscosity of slurry, defined by Eq. (9)	[kg/(m·s)]
ν	= kinematic viscosity of liquid	[m ² /s]
ν_m	= kinematic viscosity of slurry (= μ_m / ρ_m)	[m ² /s]
ρ_m	= density of slurry, defined by Eq. (10)	[kg/m ³]
ρ_p	= density of active particles	[kg/m ³]
ρ'_p	= density of inert particles	[kg/m ³]
ρ_w	= density of liquid	[kg/m ³]

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ENHANCED PERMEATION OF HYDROCARBONS THROUGH LIQUID MEMBRANES BY LIPOPHILIC SURFACTANTS

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Key Words: Extraction, Liquid Membrane, Selective Permeation, Enhanced Permeation, Lipophilic Surfactant, Hydrocarbon Separation, Emulsion

Batch extraction experiments on liquid membrane permeation were conducted in a stirring vessel to separate aromatics from five-component hydrocarbon mixtures in the presence of lipophilic surfactants added in solvent phase. The lipophilic surfactants served to make emulsion drops smaller and enhanced the permeation rates of each permeate greatly. As the results of the use of a lipophilic surfactant, both a benzene yield of 98% and a separation factor of 210 for benzene in reference to *n*-hexane were obtained at 1.5 minutes.

Introduction

Separation of hydrocarbon mixtures into their component parts by liquid membrane permeation (LMP) is a challenging theme for the petroleum refinery industry as well as the chemical industry because of the considerable reduction of energy requirements that may be achieved. In this operation, oil-in-water type (O/W) emulsion is prepared by encapsulating hydrocarbon feed with an aqueous medium containing a selected hydrophilic surfactant. The emulsion is subsequently dispersed in a solvent, which results in the formation of (O/W)/O type emulsion. More-soluble hydrocarbons in the feed-oil

droplets permeate through the aqueous medium and they leave less-permeable components in the feed-oil droplets. After the desired separation is achieved, the (O/W)/O emulsion should be settled to separate it into an O/W emulsion and a solvent phase. The recovered O/W emulsion should subsequently be demulsified by conventional means to get raffinate and aqueous surfactant solution to be recycled. Flash or simple distillation can be applied to obtain permeates from the solvent phase.

Separation of hydrocarbons by LMP was first developed by N. N. Li,⁴⁾ who demonstrated the highly selective nature of water membranes. Several other workers^{1-3,5,7)} have studied his findings further and have suggested the practical utilization of LMP on an industrial scale. But there are some problems in this

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