PARTICLE-TO-LIQUID MASS TRANSFER IN GAS-LIQUID-SOLID FLUIDIZATION

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Mass transfer from solid particles to liquid in gas-liquid-solid fluidization was experimentally investigated by means of an electrochemical method with 7–9 particle electrodes fixed at various radial positions in a column of 82.9 mm i.d. Supplementary measurements were also conducted for the solid-to-liquid mass transfer in single liquid flow, gas-liquid two-phase upflow and liquid-solid fluidization.

The effects of particle size and liquid and gas flow rates on the solid-to-liquid mass transfer coefficient, k_s, in all the systems were investigated and the mass transfer coefficient was well correlated by the following equation proposed by Ohashi *et al.* for k_s in a packed bed with single liquid flow.

$$Sh_s = 2 + 0.51(E^{1/3}d_p^{4/3}/\nu_l)^{0.60}Sc^{1/3}$$

where E is the energy dissipation rate per unit mass of liquid, d_p the particle diameter, ν_l the kinematic viscosity of liquid, Sh_s the Sherwood number and Sc the Schmidt number. The value of E was expressed by the sum of the kinetic energy supply terms due to viscous drag force across a particle and to gas agitation.

Introduction

Gas-liquid-solid fluidized beds have been used for various industrial chemical processes. The performance of the three-phase fluidized-bed reactor for such processes may depend on the rate of heat and mass transfer between solid particles and a liquid.

Many works have been reported on solid-to-liquid mass transfer in liquid-solid fluidized beds by using various measurement techniques, involving dissolution of solid into a liquid stream,[4,8,17,34] ion exchange[15,27] and adsorption.[9] Riba and Couderc[29] measured the mass transfer coefficient k_s around an active particle immersed in a fluidized bed of inert particles of the same diameter as the active particle. They concluded that the behavior of k_s on a particle fixed in a fluidized bed was similar to that on freely fluidized particles. Kikuchi *et al.*[13] reanalyzed solid-to-liquid mass transfer data for liquid-solid fluidized beds from many literature sources and concluded that the values of k_s could be correlated by the equations proposed by Ohashi *et al.*[24] This correlation method includes the energy dissipation rate per unit mass of liquid as a primary correlation parameter derived from Kolmogoroff's theory of local isotropic turbulence.[10]

Only a little research has been done on solid-liquid mass transfer in gas-liquid-solid fluidized beds.[2,22,26] Arters and Fan[2] and Prakash *et al.*[26] measured k_s for three-phase fluidized beds using a dissolution method. Nikov and Delmas[22] measured k_s on a fixed particle immersed in a fluidized bed of inert particles having the same diameter as that of the active one using an electrochemical method, and they showed that the mass transfer on the fixed particles was very similar to that on the freely fluidized particles. Also, these investigators independently proposed empirical correlation equations based on their own experimental data.

In the present study, k_s was measured in a three-phase fluidized bed by an electrochemical technique with 7–9 particle electrodes fixed at five radial positions in the column. Supplementary measurements of k_s were also carried out for various flow systems including single liquid flow, gas-liquid two-phase upflow and liquid-solid fluidized bed. The effects of particle size, solid concentration and gas and liquid flow rates on k_s were examined. An attempt was made to correlate k_s in a unified formula applicable to various liquid-solid and gas-liquid-solid flow systems in terms of specific power group.

1. Experimental

Figure 1 is a schematic diagram of the experimental apparatus. The column was composed of three sections in series: the calming section, the test section

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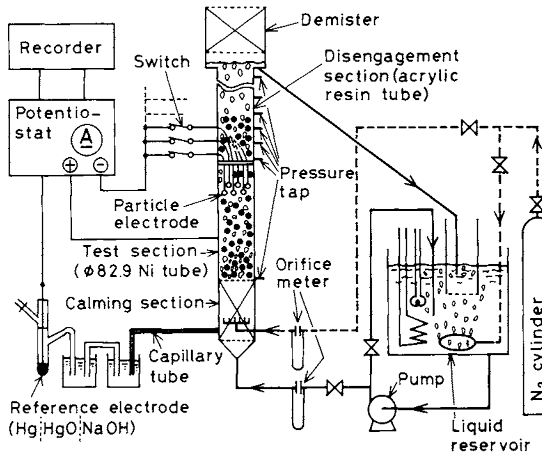

Fig. 1. Schematic diagram of experimental apparatus

and the disengagement section. The calming and disengagement sections, made of transparent acrylic resin tubes of 84 mm i.d., were 0.3 and 1.0 m in length, respectively. The calming section, with a conical-shaped bottom, was equipped with a gas injection nozzle at the bottom and was packed with 5-mm Raschig rings. The test section, used as a counter electrode to particle electrodes, was made of a nickel tube of 82.9 mm i.d. and 0.564 m in length.

As shown in **Fig. 2**, nickel-plated spheres, used as active particles for measuring the mass transfer coefficient, were fixed at radial positions of 0, 10, 20, 30 and 35 mm in a fluidized bed of electrically inert glass beads with a density of 2500 kg/m^3. The size of the active spheres was nearly equal to that of the fluidized particles; that is, active spheres with a diameter of 3.4 or 5.0 mm were used in a bed of inert particles with a diameter of 3.1 or 4.8 mm, correspondingly. As shown in Fig. 1, the fixed-particle assembly was inserted downward into the test section. The bed support, consisting of 60- and 12-mesh stainless steel screens, was placed between the calming and test sections.

The diffusion-controlled reduction of ferricyanide ion supported by large excess of caustic soda on the particle electrode was used to measure the mass transfer coefficient from particle to liquid, k_s. The value of k_s is given by the following equation:[18]

$$k_s = I/(n_e F A_s C_b) \tag{1}$$

where I is the limiting current, n_e the valence change in the electrode reaction, F the Faraday constant, A_s the area of electrode surface and C_b the bulk concentration of the ionic transport species. The properties of the electrolyte solution used are given in **Table 1**. Nitrogen was used as the gas phase.

The liquid and gas flow rates for gas-liquid-solid systems, shown in **Table 2**, range from a packed-bed state to a fluidized-bed state. The variation in static

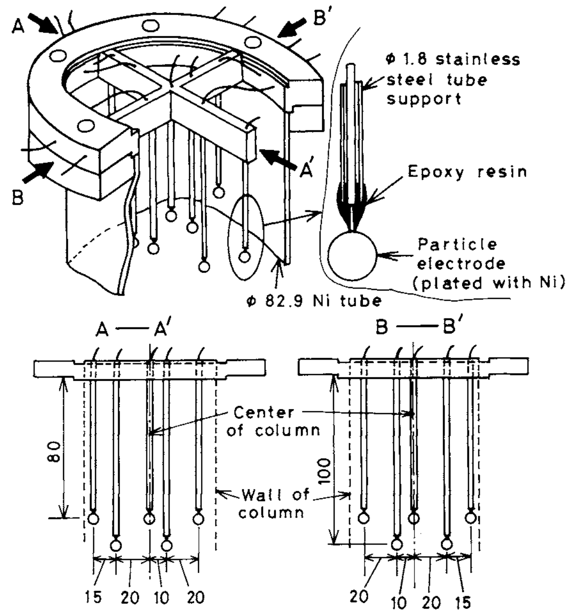

Fig. 2. Assembly of fixed particle electrode

Table 1. Properties of electrolyte solution used

Concentration of $K_3Fe(CN)_6$	1×10^{-3} mol/l*
Concentration of $K_4Fe(CN)_6$	1×10^{-3} mol/l*
Concentration of NaOH	0.2 mol/l
Viscosity	1.05×10^{-3} Pa·s
Density	1006 kg/m^3
Diffusivity of $Fe(CN)_6^{3-}$	6.97×10^{-10} m^2/s**
Schmidt number	1500
Temperature	293.2 ± 0.2 K

* Representative values.
** Estimated by the equation of Eisenberg *et al.*[6]

Table 2. Experimental conditions for fluidization systems

d_p [mm]	Mesh size [mesh]	U_l [cm/s]	U_g [cm/s]	n [—]	U_i [cm/s]
3.1	6–7	0.5–17.0	0–30.0	2.39	26.0
4.8	3.5–5	0.5–17.0	0–30.0	2.39	34.2

pressure along the column was measured by use of manometers connected to pressure taps on the column wall. The surface level of the three-phase bed was always higher than the exit of the test section by over twice the column diameter. The individual phase holdups in the fluidized bed were then calculated from the pressure drop across the bed, the bed height and the mass of solid in the column.[3,12,14] The value of liquid holdup in the liquid-solid fluidized beds was expressed well by the equation of Richardson and Zaki,[30] whose parameters, n and U_i, are shown in Table 2. The behavior of individual phase holdups in the three-phase fluidized beds were similar to those reported by various authors.[3,12,21] The gas holdup in the packed bed was estimated by the correlation of Achwal and Stepanek[1] and that of Stiegel and Shah.[33] Supplementary measurements of k_s were also carried out in a single liquid flow and a gas-liquid concurrent upflow.

2. Experimental Results and Discussion

2.1 Variation of solid-to-liquid mass transfer coefficient with radial position

Figure 3 shows the variation of solid-to-liquid mass transfer coefficient, $k_{s,r}$, with radial distance in the gas-liquid two-phase flow and three-phase fluidized bed systems. It is seen that the value of $k_{s,r}$ is almost constant in radial direction in the gas-liquid two-phase flow, but in the three-phase fluidized bed the value of $k_{s,r}$ near the wall region is about 20% smaller than that in the core region at high values of U_g. This may be due to a lower population of gas bubbles and suppression of turbulence by the presence of solid particles in the near-wall region in the three-phase fluidized bed.

In summary, however, the variation of k_s with radial position appears to be small except in the region close to the wall in gas-liquid and gas-liquid-solid flow systems, even though a considerable radial variation of gas holdup may exist in these multiphase flow systems.[20,35)]

2.2 Behavior of solid-to-liquid mass transfer coefficient, k_s

Figure 4 shows the variation of k_s with U_l in a single liquid flow and that in liquid-solid systems including a liquid-solid fluidized bed and a packed bed with U_l less than the incipient fluidization velocity. Here, the value of k_s is the mean of $k_{s,r}$ for all the active particles. The values of k_s in the single liquid flow increase with increasing U_l and with decreasing d_p, showing good agreement with values estimated from the equation of Ranz and Marshall.[28)] In the packed-bed regime of the liquid-solid system the value of k_s increases with increasing U_l and with decreasing d_p, and is much larger than that in the single liquid flow at a corresponding value of U_l. In the fluidized bed regime, however, the value of k_s becomes insensitive to liquid flow rate until the bed voidage reaches unity. It is also seen that the effect of d_p on k_s in the liquid-solid fluidized beds appears to be negligible.

Figure 5 shows the values of k_s in the gas-liquid concurrent upflow and in the gas-liquid-solid system. It is seen that the value of k_s in the gas-liquid two-phase flow increases gradually with increasing U_g, being approximately proportional to $U_g^{0.2}$. But the effect of U_l on k_s is generally small. In the gas-liquid-solid system, the value of k_s increases with increasing U_g and approaches the value in the gas-liquid two-phase flow at a high value of gas flow rate. However, in the packed-bed regime of the system at a small liquid flow rate (e.g., $U_l = 0.5$ cm/s) and at high gas flow rates ($U_g > 10$ cm/s), the value of k_s decreases with increasing U_g. In this operational condition,

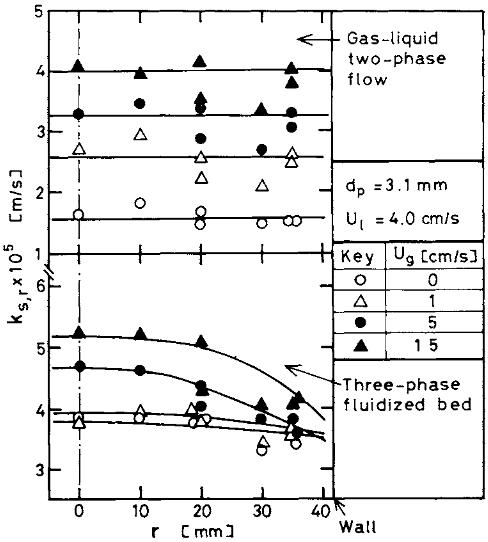

Fig. 3. Radial variation of solid-to-liquid mass transfer coefficient in a gas-liquid two-phase flow and in a three-phase fluidized bed

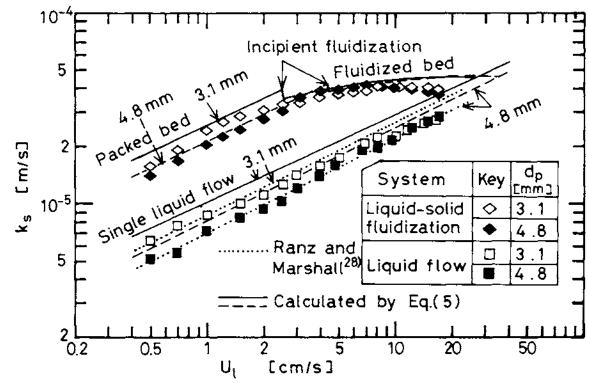

Fig. 4. Variation of k_s with U_l in a liquid-solid system and in a single liquid flow

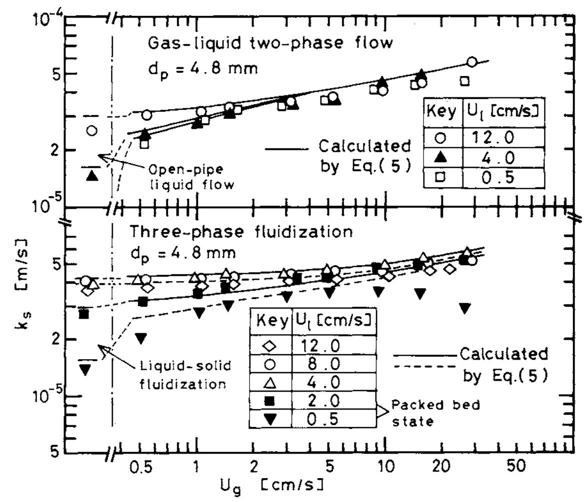

Fig. 5. Effects of U_g and U_l on k_s in a gas-liquid two-phase flow and in a three-phase fluidized bed

large gas slugs ascend frequently and entirely cover the active particles, so that the effective liquid-solid contact area may decrease, thus decreasing the overall

Table 3. Equations for evaluation of pressure drop across a particle in various flow systems

System	Equations for ΔP_d		Investigators
Packed bed with liquid flow	$\Delta P_d=(150\mu_l U_l/d_p)(1-\varepsilon_l)^2/\varepsilon_l^3 + 1.75\rho_l U_l^2(1-\varepsilon_l)/\varepsilon_l^3$ (Ergun[7])	(T-1)	Ohashi *et al.*[24]
	$\Delta P_d=100(1-\varepsilon_l)C_{D0}\rho_l U_l^2/2$ (Shirai[32])	(T-2)	
Packed bed with gas-liquid concurrent upflow	ΔP_d is the dynamic pressure drop corrected for the liquid head from the static pressure drop.		
Single particle in liquid flow	$\Delta P_d=C_{Dr}\rho_l U_r^2/2$ where U_r is relative velocity.	(T-3)	Ohashi *et al.*[24]
Particles in fluidization	$\Delta P_d=\varepsilon_l C_{Di}\rho_l U_i^2/2$ (Ishii and Zuber[11])	(T-4)	Kikuchi *et al.*[13]

mass transfer rate. The influence of U_l on k_s in the three-phase fluidized bed is very small as is the case in the liquid-solid fluidized bed.

It is noted that at a small liquid flow rate the value of k_s increases sharply when a small volume of gas is introduced into the single liquid flow or the liquid-solid system. On the other hand, at a high liquid flow rate in either flow system the effect of introducing a gas on k_s is small. Such behavior of k_s may be explained as follows. At a small liquid flow rate, the turbulence in liquid phase is intensified much more by gas injection than by the bulk liquid flow; hence, the rate of solid-to-liquid mass transfer may be mainly determined by the gas agitation. At a high liquid flow rate, however, the turbulence generated by the bulk liquid flow across the solid particles may play a dominant role in determining the mass transfer rate.

2.3 Correlation of k_s

In correlating the mass transfer coefficient, one can write the following relationship:

$$Sh=f(Re, Sc) \tag{2}$$

It seems from the results of the previous section that the mass transfer coefficient may increase with an increase in the extent of turbulence in the liquid phase generated by gas agitation as well as by bulk liquid flow across the solid particle. To account for such a turbulence effect on the mass transfer coefficient of various multiphase flow systems, many investigators applied Kolmogoroff's theory of local isotropic turbulence,[10] which relates the above Reynolds number to an easily measurable macroscopic quantity, the "specific power group":

$$Re=c(Ed_p^4/\nu_l^3)^m \tag{3}$$

where E is the energy dissipation rate per unit mass of liquid and ν_l is the kinematic viscosity of liquid. The value of m is 1/3 for $d_p \gg \eta$ and 1/2 for $d_p \ll \eta$, where η is the length scale of the smallest eddies in the isotropic turbulent flow field.

Ohashi *et al.*[24] reanalysed many published experimental data on k_s for a single particle fixed in single liquid flow and for packed particles with liquid flow and proposed the following two equations over a wide range of operational conditions:

for fixed single particles in single liquid flow

$$Sh_s=2+0.59(E^{1/3}d_p^{4/3}/\nu_l)^{0.57}Sc^{1/3} \tag{4}$$

and for packed particles

$$Sh_s=2+0.51(E^{1/3}d_p^{4/3}/\nu_l)^{0.60}Sc^{1/3} \tag{5}$$

Actually, the difference between these two equations can be ignored in the range of $1<E^{1/3}d_p^{4/3}/\nu_l<10^4$ allowing for the scatter of the experimental data. Ohashi *et al.*[24] also mentioned that the two equations are essentially identical with the correlations for k_s in suspended bubble columns[31] and stirred tanks.[16] Kikuchi *et al.*[13] confirmed that Eqs. (4) and (5) were also applicable to the data of k_s in liquid-solid fluidized beds obtained by many investigators.

The quantity of energy dissipated in the liquid phase can be expressed by the total kinetic energy supplied to the liquid phase. In a gas-liquid-solid system, the total kinetic energy supplied to the liquid phase is generated from drag force across a particle, gas agitation and wall shear stress. Consequently, the value of E can be expressed by the following relation in terms of the three energy supply sources:

$$E=(\Delta P_d/d_p)U_l/(\varepsilon_l\rho_l)+U_g g+(\Delta P_w/H)U_l/(\varepsilon_l\rho_l) \tag{6}$$

where ΔP_d is the pressure drop across a particle of diameter d_p, and ΔP_w the pressure drop due to wall friction across the bed height H. The equations for evaluating ΔP_d in various liquid-solid and gas-liquid-solid systems are listed in **Table 3**. The last term of the right-hand side of Eq. (6) can be generally ignored except in the case of a solid transport system.[23,25]

Typical values of E for various flow systems calculated from the equations for ΔP_d given in Table 3 are shown in **Fig. 6**. The value of E in the gas-liquid two-phase flow is of the same order as in the liquid-solid fluidized bed, whereas the value of E in the packed bed with liquid flow at U_l less than 1 cm/s is one order of magnitude smaller than that in the gas-liquid two-phase flow. The curve for E in the liquid-solid fluidized bed lies between those for E in the packed bed and in the single liquid flow across a particle. The dependence of E on d_p and U_l is much smaller in the liquid-solid fluidized bed than in the packed bed or in the single liquid flow.

The value of E in the three-phase fluidized bed can be given simply by the sum of the values of E in the liquid-solid fluidized bed and in the gas-liquid two-phase flow, since for fluidization systems substitution of Eq. (T-4) for ΔP_d in Table 3 into Eq. (6) eliminates the unknown parameter ε_l, allowing the first term of the right-hand side of Eq. (6) to be a function of U_l alone. The value of E in the packed bed with gas-liquid upflow is approximately given by the sum of the values of E in the gas-liquid two-phase flow and in the packed bed with single liquid flow. Note that in the real calculation of E for the packed bed with gas-liquid upflow the value of ΔP_d was obtained from the experimental value of pressure drop corrected by liquid head. It is also noted that the behavior of E well reflects that of k_s for various multiphase flow systems, indicating that the value of E may be employed as the key parameter for correlating k_s.

Figure 7 shows the plot of the experimental results of the present study based on the energy dissipation concept. It is seen that all the experimental values of

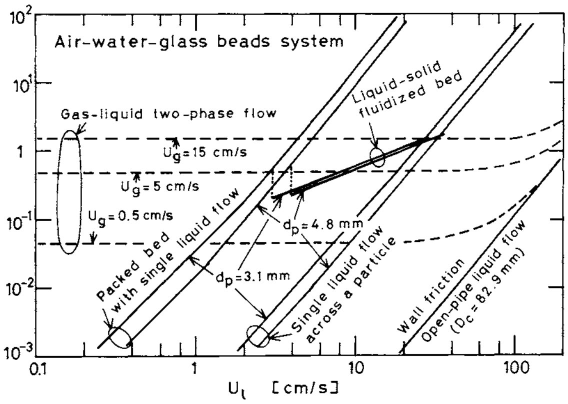

Fig. 6. Typical values of E in various gas-liquid and liquid-solid flow systems

k_s except those in slugging flow regime in the concurrent packed bed are well correlated by Eqs. (4) and (5). The coefficient of variation of these correlation equations is about 0.26 for 205 data points. The values of k_s estimated by Eq. (5) are shown in Figs. 4 and 5, indicating good agreement with those observed.

This result may indicate that the specific power group can be chosen as a singular parameter to evaluate the hydrodynamic effect on the solid-to-liquid mass transfer in gas-liquid-solid systems as well as in liquid-solid systems.

Also shown in Fig. 7 are the values of Sh_s in a suspended bubble column estimated by the correlation of Sano *et al.*[31] It is seen that the value of Sh_s for the suspended bubble column is somewhat larger than the experimental values in the three-phase fluidized beds in this work. The difference between the two values may be due to the difference in sizes of particles used and the differences in statistical properties of liquid-phase turbulence in the two systems.

2.4 Comparison of correlations for k_s

Arters and Fan[2] measured k_s in liquid-solid and three-phase fluidized beds using dissolution of benzoic acid particles ($\rho_s = 1300\,\text{kg/m}^3$) and found that the value of k_s increased significantly with increasing U_g in the three-phase fluidized bed by up to 150% over the value in the liquid-solid fluidized bed. Prakash *et al.*[26] measured k_s using a few glass beads coated with benzoic acid in a three-phase fluidized bed with glass beads ($\rho_s = 2520\,\text{kg/m}^3$) and found that the magnitude of increase in k_s with increasing U_g is not so large as that reported by Arters and Fan.[2] Nikov and Delmas[22] measured k_s by the electrochemical method, examined the influence of several operating parameters on k_s and concluded that the lighter the density of solid is, the more steeply the value of k_s increases with increasing U_g.

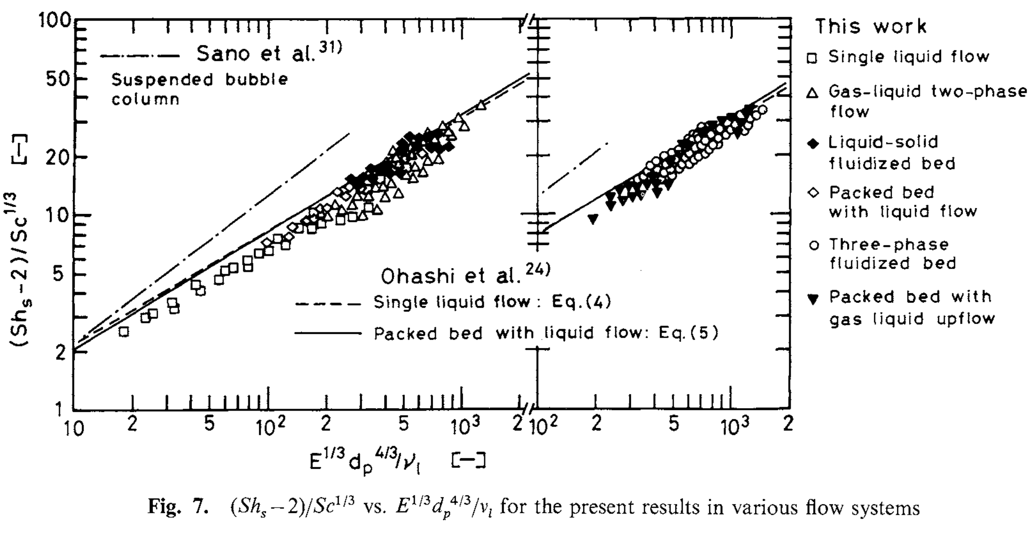

Fig. 7. $(Sh_s-2)/Sc^{1/3}$ vs. $E^{1/3}d_p^{4/3}/\nu_l$ for the present results in various flow systems

The values of k_s estimated by the correlation of Arters and Fan[2] are much higher (by more than 100%) than those observed in the present work. This discrepancy between the observed and the estimated values of k_s is due to the fact that the density of the particles used by Arters and Fan[2] is much smaller than that in the present work. The values of k_s predicted by the correlation of Prakash *et al.*[26] agree fairly well with those observed in the present work except in a region of high liquid flow rates, where the estimated values are about 60% larger than the observed ones. From the viewpoint of the energy dissipation concept, for the case of a light-particle system as used by Arters and Fan[2] the energy dissipation rate due to drag force across a particle is relatively small in comparison with that due to gas agitation, so that the mass transfer rate can be dominantly affected by the intensity of gas agitation.

As shown in **Fig. 8**, the values of k_s observed by Arters and Fan,[2] Prakash *et al.*[26] and Nikov and Delmas[22] are well correlated by Eq. (5) with the aid of the value of E from Eq. (6). It is apparent that the enhancement effect of $(\rho_s - \rho_l)$ on k_s as observed by Nikov and Delmas[22] is well represented by the energy supply due to drag force across the particle involved in Eq. (6).

In Fig. 8, the data of k_s in packed beds with gas-liquid concurrent upflow measured by various workers[5,19,36] are plotted, based on the energy dissipation rate from Eq. (6). It is shown that the data of k_s in the packed bed with gas-liquid concurrent upflow are also correlated well by Eq. (5).

Concluding Remarks

The value of the solid-to-liquid mass transfer coefficient, k_s, for the gas-liquid-solid fluidized bed increases with increasing gas flow rate from the value of k_s in the liquid-solid fluidized bed and appears to approach the value in the gas-liquid two-phase flow at a high value of gas flow rate. The effects of particle diameter and liquid flow rate on k_s are small in both the liquid-solid and the three-phase fluidized beds. The variation of k_s with radial position is generally small except in the region close to the wall in both the gas-liquid and the gas-liquid-solid systems.

The data of k_s for the gas-liquid two-phase upflow, the three-phase fluidized bed and the packed bed with gas-liquid concurrent upflow are correlated well by Eqs. (4) and (5) proposed by Ohashi *et al.*[24] based on the energy dissipation concept. Here, the energy dissipation rate is evaluated by Eq. (6) as the sum of the energy supply terms due to drag force across a particle and that due to gas agitation.

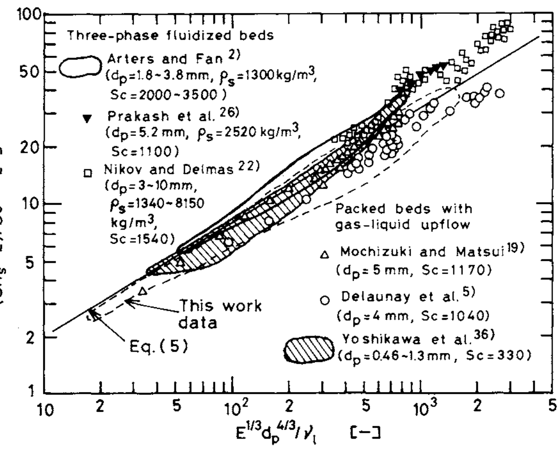

Fig. 8. Comparison of Eq. (5) with published data on k_s in three-phase fluidized beds and in packed beds with gas-liquid upflow

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Nomenclature

A_s	= area of electrode surface	[m^2]
C_b	= bulk concentration of transport species	[mol/m^3]
C_{Di}	= drag coefficient for a single particle flowing at a rate of U_i against liquid	[—]
C_{Dr}	= drag coefficient for a single particle flowing at a rate of U_r against liquid	[—]
C_{D0}	= drag coefficient for a single particle fixed in liquid flowing at a rate of U_l	[—]
c	= constant in Eq. (3)	[—]
d_p	= particle diameter	[m]
E	= energy dissipation rate per unit mass of liquid	[m^2/s^3]
F	= Faraday constant	[C/mol]
g	= gravitational acceleration	[m/s^2]
H	= bed height	[m]
I	= limiting current	[A]
k_s	= mass transfer coefficient between solid particles and liquid	[m/s]
$k_{s,r}$	= k_s at a radial position of r	[m/s]
m	= exponent in Eq. (3)	[—]
n	= exponent in Richardson–Zaki equation	[—]
n_e	= valence change in electrode reaction	[—]
ΔP_d	= pressure drop across single particle	[Pa]
ΔP_w	= pressure drop across a bed height due to wall friction	[Pa]
Re	= Reynolds number	[—]
r	= radial distance	[m]
Sh	= Sherwood number	[—]
Sh_s	= Sherwood number concerning solid-to-liquid mass transfer	[—]
Sc	= Schmidt number	[—]
U_g	= superficial gas velocity	[m/s]
U_i	= apparent liquid velocity at unit voidage in Richardson–Zaki equation	[m/s]
U_l	= superficial liquid velocity	[m/s]
U_r	= relative velocity between solid and liquid	[m/s]
ε_l	= liquid holdup	[—]
η	= length scale of smallest eddies in isotropic turbulence	[m]
μ_l	= liquid viscosity	[Pa·s]
ν_l	= kinematic viscosity of liquid	[m^2/s]

ρ_l	= liquid density	[kg/m^3]
ρ_s	= solid density	[kg/m^3]

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