WALL-TO-LIQUID MASS TRANSFER IN PACKED AND FLUIDIZED BEDS WITH GAS-LIQUID CONCURRENT UPFLOW

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Key Words: Mass Transfer, Chemical Reactor, Mass Transfer Coefficient, Three-Phase Fluidization, Packed Bed, Multiphase Flow, Energy Dissipation Rate, Heat Transfer, Analogy

The wall-to-liquid mass transfer coefficient, k_w, was measured for both surfaces of a coaxially immersed tube and a column wall in packed and fluidized bed systems with gas-liquid concurrent upflow. Supplementary measurements of k_w were carried out in openpipe liquid flow, gas-liquid two-phase upflow and packed and fluidized beds with single liquid flow.

The value of k_w in the three-phase fluidized bed increased with increasing gas flow rate, deviating from the value in the liquid-solid fluidized bed and approaching the value in the gas-liquid two-phase upflow, while it passed through a maximum value with respect to the liquid flow rate. The value of k_w in the packed-bed operation increased with increasing liquid flow rate and with increasing gas flow rate. The values of k_w for the inner tube and the column wall were shown to agree with each other.

The values of k_w in all the gas-liquid, liquid-solid and gas-liquid-solid systems examined were correlated well by a unified equation in terms of the energy dissipation rate per unit mass of liquid.

An analogy existed between wall heat transfer and wall mass transfer in all the multiphase flow systems examined.

Introduction

Gas-liquid-solid fluidized and packed beds can be commonly used as three-phase reactors and contactors in various chemical processes. When these beds are utilized as electrochemical reactors, the rate of wall-to-liquid mass transfer is an important factor in the design and operation of such reactors. The wall-to-liquid mass transfer characteristic is also important since it usually relates to the wall-to-bed heat transfer characteristic.

There are many studies on the wall-to-liquid mass transfer in a liquid-solid fluidized bed[4,5,9,14] and a packed bed with single liquid flow.[14,16] It may be thought that the mass transfer rate from the wall surface is significantly increased by introducing gas phase into a liquid-solid fluidized bed or a packed bed with single liquid flow because of gas-phase agitation. Only a little work[9] has, however, been done on wall-to-liquid mass transfer in the three-phase fluidized bed and the packed bed with gas-liquid concurrent flow.

Wall-to-liquid mass transfer measurements have been made over surfaces of various shapes in liquid-solid fluidized beds—the column wall surface,[4,14] rectangular bed surface[9] and immersed tube surface,[5] for example. However, the effect of the physical geometry of the transfer surface on the wall-to-liquid mass transfer coefficient, k_w, has not been clear.

In a previous paper[11] it was confirmed that the analogy between heat and mass transfer on the column wall holds for liquid-solid fluidized beds. The applicability of that analogy to three-phase fluidized beds was also suggested.[12]

In the present study, wall-to-liquid mass transfer experiments were carried out in fluidized and packed bed systems with single liquid flow and with concurrent gas-liquid upflow for two wall geometries: a coaxially submerged tube wall and a column wall. Supplementary experiments were carried out in a gas-liquid concurrent upflow system. The influences of operating conditions and characteristic size of the wall on the behavior of k_w were also investigated. Based on the experimental results a unified correlation equation for k_w was developed and is applicable to various multiphase flow systems. This equation was then tested to correlate the published data on k_w obtained for transfer surfaces of various shapes. An analogy between wall mass transfer and wall heat transfer in various multiphase flow systems was also considered.

1. Experimental Apparatus and Procedure

The column and the gas and liquid flow systems

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used in this study are essentially the same as those used for measuring the solid-to-liquid mass transfer coefficient elsewhere.[3] **Figure 1** shows a schematic diagram of the experimental setup for measuring the wall mass transfer coefficient. The column was composed of three sections in series: the calming section, the test section and the disengagement section, all having an almost identical inside diameter. The test section, with a total length of 0.564 m, was constructed of six nickel electrodes of 82.9 mm i.d. and 3 mm thickness connected in series and insulated electrically from each other by thin polyethylene sheets with a thickness of 0.03 mm. The surface of each electrode was polished to mirror smoothness by means of finest-grain sandpaper. The length of each electrode is given in **Table 1**.

One of three counter electrodes with diameters of 3, 10 and 25 mm was coaxially inserted in the test section. The electrode of 3 mm diameter was made of nickel rod, while the other two were constructed of four nickel-plated copper tubes connected in series and insulated electrically from each other by thin acrylic resin sheets with a thickness of 0.5 mm. The total length of the inner electrodes assembly with a diameter of 10 or 25 mm was almost equal to that of the column wall electrodes assembly. The dimensions of all the inner electrodes used are given in Table 1.

An electrochemical method for measuring the particle-to-liquid mass transfer coefficient[3] was used to determine the wall-to-liquid mass transfer coefficient. To measure the wall-to-liquid mass transfer coefficient on the column wall, the column wall electrodes were used as the cathode and the inner tube electrodes were used as the anode, as shown in Fig. 1. The electric circuit was designed to obtain the limiting current for any combination of cathodes. In the figures, for example, only No. 5 cathode is connected to the measuring circuit. All cathodes were held at the same potential to eliminate disturbance of the electric potential field and to minimize the entrance effect of the concentration boundary layer on the measuring electrode.

In the case of mass transfer measurement on the inner tube, the inner-tube electrodes were used as cathode and the column-wall electrodes as anode. The procedure for measuring the mass transfer coefficient over the inner tube is identical to that for mass transfer measurement on the column wall.

The liquid used was an aqueous solution of a mixture of potassium ferricyanide and potassium ferrocyanide in large excess of sodium hydroxide. The physicochemical properties of the liquid were as follows: density = 1006 kg/m^3, viscosity = 1.05×10^{-3} Pa·s and Schmidt number = 1500. The concentration of ferricyanide ion was determined for each run of the mass transfer measurement on the column

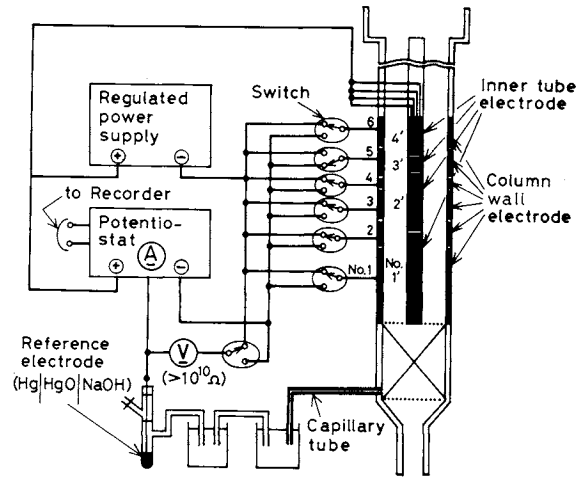


Fig. 1. Experimental setup for measuring wall-to-liquid mass transfer coefficient

Table 1. Dimensions of column wall and inner tube electrodes

Column wall electrode	No.	1	2	3	4	5	6
	L [mm]	240	80	80	4	80	80
Inner tube electrode	D_i = 10 mm	No.		1′	2′	3′	4′
		L [mm]		252	174	13	119
	D_i = 25 mm	No.		1′	2′	3′	4′
		L [mm]		250	167	13	132
	D_i = 3 mm	Entire rod was used as single electrode (L = 2 m).					

Table 2. Size of glass beads and experimental conditions

Fluidization systems

d_p [mm]	Mesh size [mesh]	U_l [cm/s]	U_g [cm/s]	n [—]	U_i [cm/s]
0.77	20–24	0.5–12	0–17	3.24	12.5
1.1	14–16	0.5–13	0–17	2.91	14.2
3.1	6–7	0.5–20	0–17	2.39	26.0
4.8	3.5–5	0.5–20	0–17	2.39	34.2

Packed bed systems

d_p [mm]	Mesh size [mesh]	U_l [cm/s]	U_g [cm/s]
1.1	14–16	0.5– 6	0
3.1	6–7	0.5–15	0–30
6.2	3–3.5	0.5–17	0–30

wall by means of iodometry, since it decreased gradually from run to run because of the overpotential on the anode. Nitrogen was used as the gas phase. Glass beads with a density of 2500 kg/m^3 were used as the solid phase. The size of solids used and the ranges of gas and liquid flow rates are shown in **Table 2**.

In fluidization, the bed surface was always adjusted to a level of about 0.2 to 0.5 m above the outlet of the test section. The variation in static pressure along the

column was measured by use of liquid manometers. The individual phase holdups were calculated from the pressure drop across the bed, the bed height and the mass of the fluidized solids.[10] The liquid holdup in the liquid-solid fluidized bed was well correlated by the equation of Richardson and Zaki.[13] The parameters of the equation, n and U_i, determined experimentally in this study, are also shown in Table 2. The effects of operating variables such as particle size, liquid flow rate and gas flow rate on the individual phase holdups in three-phase fluidized beds were found to be similar to those reported in the literature.[6,10] On the other hand, in packed beds the solid particles were tightly fixed in the test section by wire mesh screens placed at the upper and lower bounds. The pressure drop across the bed was measured by means of a mercury manometer. Gas and liquid holdups in the packed beds with gas-liquid concurrent upflow were estimated by using correlation equations.[1,15]

2. Experimental Results and Discussion

2.1 Behavior of wall-to-liquid mass transfer coefficient, k_w

In the entrance region of the column ($z/D_c < 3$), the values of k_w for the column wall and the inner tube varied slightly with axial position for gas-liquid, liquid-solid and gas-liquid-solid systems. Consequently, the value of k_w on No. 5 cathode of the column-wall electrodes assembly and that on No. 2′ or 3′ cathode of the inner-tube electrodes assembly were respectively adopted as representative values for the mass transfer coefficients on the column wall and on the inner tube.

Figure 2 shows the effect of the liquid flow rate, U_l, on the values of k_w in a packed bed with single liquid flow. It is apparent that the values of k_w are proportional to the 0.6–0.62 power of U_l and increase with decreasing d_p. The difference between the values of k_w on the column wall and on the inner tube is negligible. The values of k_w estimated from the correlation of Yagi and Wakao[16] are presented by broken lines in the figure. It is seen that agreement between the values of k_w estimated from Yagi and Wakao[16] and those observed in the present work is fairly good.

Figure 3 shows the variation of k_w with the gas flow rate, U_g, in a packed bed with gas-liquid concurrent upflow. It is seen that the values of k_w increase with increase of either U_g or U_l and that the difference between the values of k_w on the inner tube and on the column wall is very small.

Figure 4 shows the behavior of k_w in liquid-solid fluidization and also that in open-pipe liquid flow for comparison. Note that in the laminar-flow region for open-pipe liquid flow the values of k_w were measured

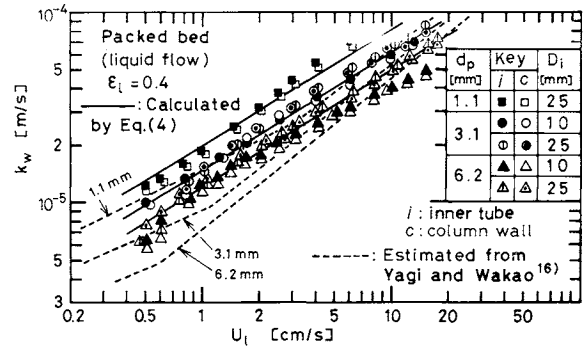


Fig. 2. Variation of k_w with U_l in packed beds with single liquid flow

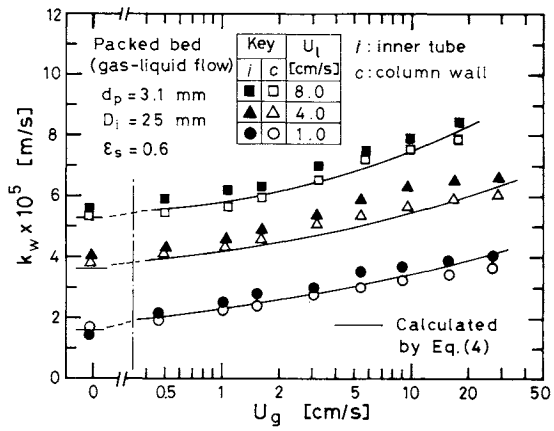


Fig. 3. Effect of U_g on k_w in a packed bed with gas-liquid concurrent upflow

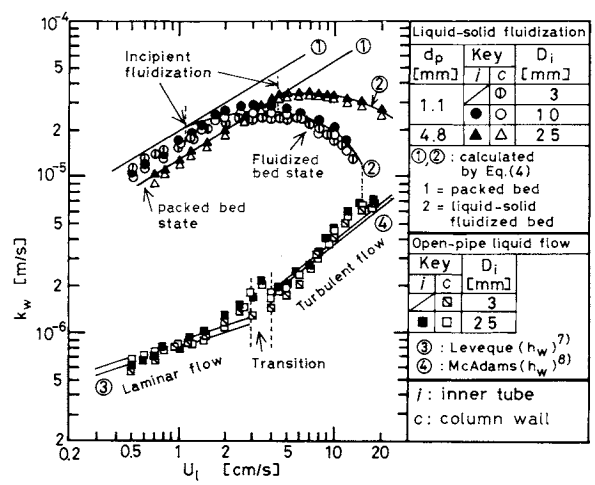


Fig. 4. Variation of k_w with U_l in a liquid-solid fluidized bed and in an open-pipe liquid flow

on the entire surface of the electrodes assembly. When the value of U_l is less than the incipient fluidization velocity, i.e., the bed is in a packed-bed state, the value of k_w increases with increasing U_l, showing good agreement with the value in the tightly packed bed given in Fig. 2. The value of k_w in the packed bed is, however, 10 times larger than that in the open-pipe liquid flow. At the onset of fluidization the value of k_w becomes insensitive to U_l, then passes through a

maximum and decreases with further increase in U_l. Finally, the value of k_w in the liquid-solid fluidized bed appears to approach that in open-pipe liquid flow. It is also seen that the value of k_w increases with increasing d_p in the fluidized bed, while the reverse trend is observed in the variation of k_w in the packed bed. The effect of tube diameter on k_w for the inner tube is small and the difference between the values of k_w on the inner tube and on the column wall is also small. It is noted that in open-pipe liquid flow the value of k_w agrees well with that estimated by the equation of Leveque[7] in the laminar-flow region and that estimated by the equation of McAdams[8] in the turbulent-flow region, on the basis of the analogy concept for heat and mass transfer.

Figure 5 shows the effect of inner tube size on k_w in gas-liquid two-phase flow and the three-phase fluidized bed. It is seen that the effect of the inner tube diameter on the values of k_w is small in general for the surfaces of both the inner tube and the column wall in these systems. The value of k_w for the inner tube of 10 mm diameter is, however, somewhat larger than that for the column wall at a high value of U_g.

Figure 6 shows the variation of k_w for the column wall with U_g at various U_l in gas-liquid two-phase flow and the three-phase fluidized bed. It is seen that in both systems the value of k_w increases gradually with increasing U_g. The value of k_w for the three-phase fluidized bed is larger than that for the gas-liquid two-phase flow at a moderate gas flow rate, but the difference between the values of k_w in gas-liquid two-phase flow and the three-phase fluidized bed becomes small at a high gas flow rate. Note that the increase of k_w due to the introduction of a small amount of gas into an open-pipe liquid flow is so great that the effect of U_l on k_w becomes negligible in gas-liquid two-phase flow. This steep increase of k_w may be due to the increase of turbulence in the liquid phase by the gas bubble agitation. On the contrary, the increase of k_w caused by introducing a small amount of gas into a liquid-solid fluidized bed is relatively small in comparison with the case of gas-liquid upflow. At low and moderate gas flow rates, the variation of k_w with U_l in the three-phase fluidized bed is similar to that in the liquid-solid fluidized bed. However, at a high gas flow rate above 10 cm/s the effect of U_l on k_w becomes small.

2.2 Correlation of k_w

From the results described in the section above, it is suggested that the behavior of k_w is similar to that of solid-to-liquid mass transfer coefficient in packed and fluidized beds and in gas-liquid two-phase flow as reported elsewhere.[3] It can be recalled that the solid-to-liquid mass transfer coefficients in various multiphase flow systems have been successfully correlated by a unified correlation by using the energy dissi-

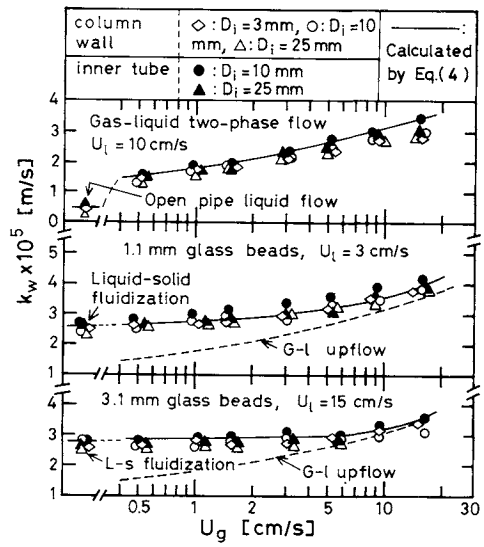


Fig. 5. Effect of inner tube size on k_w in a gas-liquid two-phase flow and in a three-phase fluidized bed

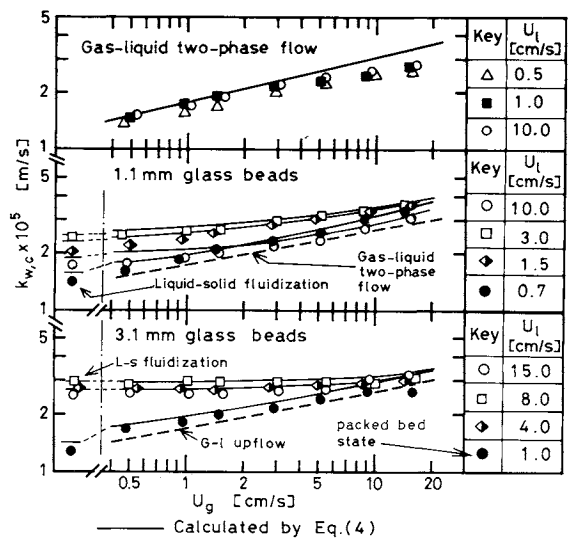


Fig. 6. Effect of U_g and U_l on k_w for the column wall in a gas-liquid two-phase flow and in a three-phase fluidized bed

pation rate per unit mass of liquid, E.[3] Along the same line of thought, an attempt was made here to correlate the Sherwood number concerning k_w in terms of the specific power group including E and the Schmidt number. In gas-liquid-solid flow systems, the average value of E across the bed may be expressed by the sum of three energy source terms as follow:

$$E=(-dP_l/dz)U_l/(\varepsilon_l\rho_l)+U_g g + (-dP_w/dz)U_l/(\varepsilon_l\rho_l) \quad (1)$$

where $(-dP_l/dz)$ is the dynamic pressure gradient in the bed. In packed-bed systems, $(-dP_l/dz)$ is identical to $\Delta P_d/d_p$, the axial pressure drop across a particle.[3] The values of $(-dP_l/dz)$ in fluidized-bed systems are given by the following equations;

for liquid-solid fluidized beds

$$(-dP_l/dz)=\varepsilon_s(\rho_s-\rho_l)g \tag{2}$$

for three-phase fluidized beds

$$(-dP_l/dz)=\{\varepsilon_s(\rho_s-\rho_l)-\varepsilon_g(\rho_l-\rho_g)\}g \tag{3}$$

On the other hand, $(-dP_w/dz)$ is the axial pressure gradient due to wall friction, which can be neglected in fluidization systems except for the highly expanded liquid-solid fluidized bed with bed porosity larger than about 0.95.

Figures 7 and **8** show the experimental results of the present study based on the energy dissipation concept. It is seen that the Sherwood numbers for k_w in all the systems can be well correlated in terms of the specific power group and the Schmidt number by a single formula:

$$Sh_w=0.13(E^{1/3}D^{4/3}/\nu_l)^{0.75}Sc^{1/3} \tag{4}$$

where $Sh_w=k_wD/D_l$ and D is the characteristic length of the mass transfer surface. (D is replaced by D_c for the column wall or D_i for the inner tube.) The coefficient of variation for Eq. (4) is 0.21 for 1230 data points. This equation shows that the geometric size of the transfer surface does not affect the correlation of the wall-to-liquid mass transfer coefficient as long as the characteristic length of the mass transfer surface is properly chosen. The values of k_w calculated by Eq. (4) are also shown in Figs. 2–6. It is seen that the calculated values of k_w agree well with observed values over a wide range of operating conditions in gas-liquid, liquid-solid and gas-liquid-solid flow systems.

This correlation method is then applied to re-analyze the literature data on k_w obtained for various geometrical shapes of mass transfer surface in liquid-solid fluidized beds.[4,5,9] The results are shown in **Fig. 9**. It is seen that the values of k_w for various geometrical surfaces in liquid-solid fluidized beds are well correlated by Eq. (4).

2.3 Applicability of the correlation based on the energy dissipation concept to wall heat transfer

In our previous papers,[11,12] wall-to-bed heat transfer phenomena in liquid-solid and three-phase fluidized beds were analyzed on the basis of a series thermal resistance model. The apparent wall heat transfer coefficients, h_w, in both kinds of fluidized beds were correlated by means of the Colburn j-factor approach and the existence of the analogy between the wall heat transfer and the wall mass transfer was confirmed for the liquid-solid fluidized bed. From these findings on the analogy between heat and mass transfer, Eq. (4) may be converted into the following form in terms of corresponding dimensionless groups for heat transfer.

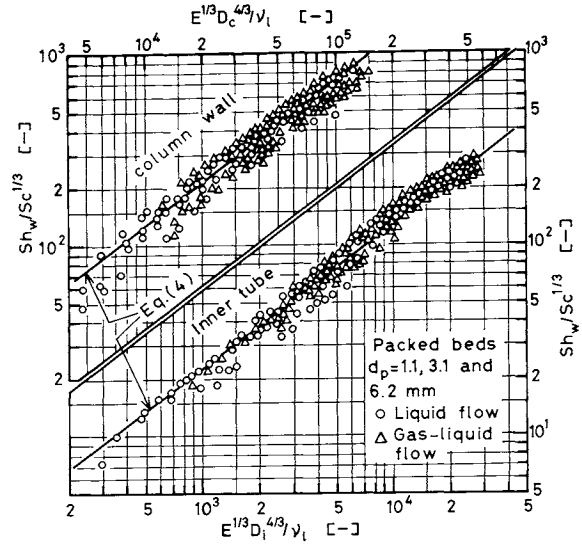


Fig. 7. $Sh_w/Sc^{1/3}$ vs. $E^{1/3}D^{4/3}/\nu_l$ in packed beds

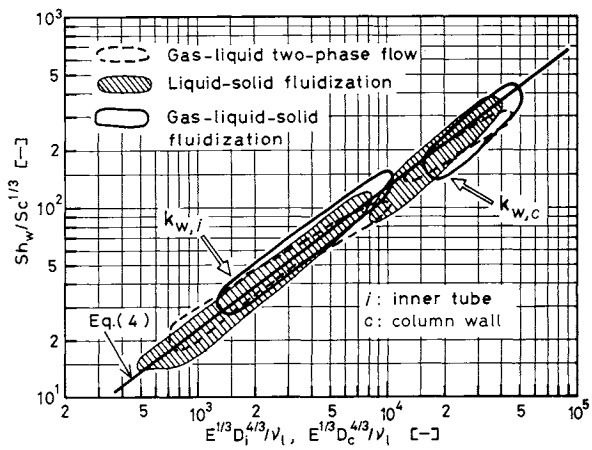


Fig. 8. $Sh_w/Sc^{1/3}$ vs. $E^{1/3}D^{4/3}/\nu_l$ in a gas-liquid two-phase flow and in fluidized beds

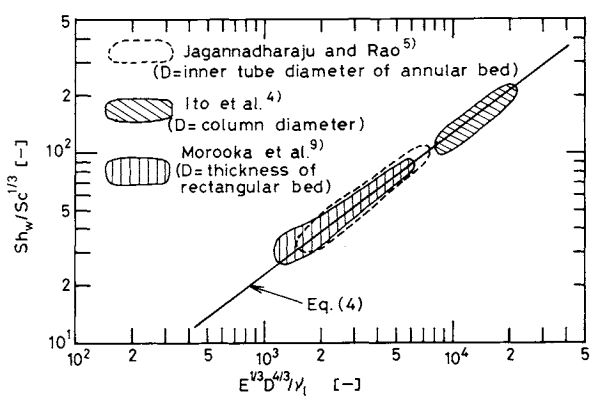


Fig. 9. Comparison of Eq. (4) with published data on k_w in liquid-solid fluidized beds

$$Nu_w=0.13(E^{1/3}D^{4/3}/\nu_l)^{0.75}Pr^{1/3} \tag{5}$$

where

$$Nu_w=h_wD/k_l \quad \text{and} \quad Pr=C_{pl}\mu_l/k_l\,.$$

The values of $Nu_w/Pr^{1/3}$ in liquid-solid and three-

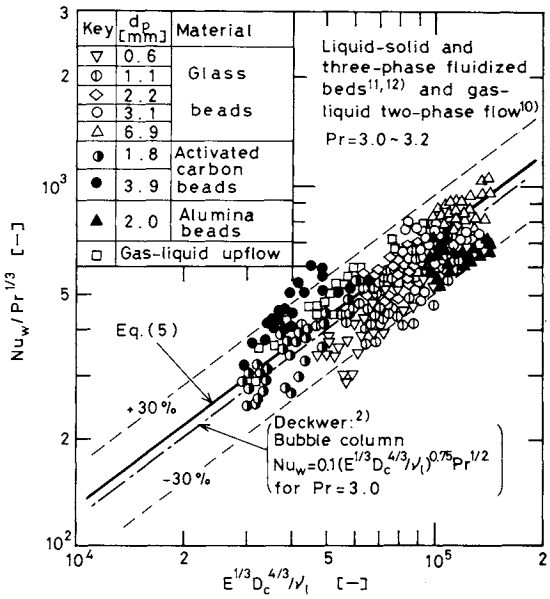


Fig. 10. Comparison of Eq. (5) with published data on h_w in a gas-liquid system and in liquid-solid and gas-liquid-solid fluidized beds

phase fluidized beds[11,12] are plotted against $E^{1/3}D^{4/3}/\nu_l$ in Fig. 10 together with the values in gas-liquid two-phase flow from the results presented elsewhere.[10] It is seen that the data of h_w in all the systems[10-12] are well correlated by Eq. (5). Consequently, it is apparent that an analogy between the wall heat transfer and the wall mass transfer also exists in the three-phase fluidized bed and in the gas-liquid two-phase flow.

On the other hand, Deckwer[2] developed a correlation for wall-to-liquid heat transfer in bubble columns based on the surface renewal theory combined with the local isotropic turbulence theory. His correlation equation expressed in terms of the specific power group is also shown in Fig. 10 at $Pr=3.0$. It is seen that the values of $Nu_w/Pr^{1/3}$ from Deckwer's correlation are very close to those calculated from Eq. (5). It is noted, however, that the modified formula of Deckwer's correlation for wall mass transfer based on the analogy concept fails to correlate the present data of k_w in gas-liquid two-phase flow because of high Sc number.

Conclusions

(1) The behavior of wall-to-liquid mass transfer coefficient, k_w, for the column wall is virtually identical with that for a vertically immersed tube in packed and fluidized bed systems with gas-liquid concurrent upflow as well as with single liquid flow.

(2) In packed-bed systems, the value of k_w increases with increasing liquid flow rate and with increasing gas flow rate. In the liquid-solid fluidized bed, however, the value of k_w shifts from that in the packed bed to that in open-pipe liquid flow passing through a maximum with increasing liquid flow rate. The value of k_w in three-phase fluidized beds progressively increases with increasing gas flow rate and approaches that in the gas-liquid two-phase upflow at a high value of gas flow rate.

(3) The values of k_w for gas-liquid two-phase flow and packed- and fluidized-bed systems with single liquid flow and with gas-liquid concurrent upflow have been correlated well by Eq. (4) on the basis of the energy dissipation concept.

(4) An analogy existed between wall mass transfer and wall heat transfer in gas-liquid two-phase flow and in liquid-solid and gas-liquid-solid fluidized beds.

Acknowledgment

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Nomenclature

C_{pl}	=	specific heat of liquid	$[J\cdot kg^{-1}\cdot K^{-1}]$
D	=	characteristic length of heat or mass transfer surface	[m]
D_c	=	column diameter	[m]
D_i	=	inner tube diameter	[m]
D_l	=	diffusivity of transfer species	$[m^2/s]$
d_p	=	particle diameter	[m]
E	=	energy dissipation rate per unit mass of liquid	$[m^2/s^3]$
g	=	gravitational acceleration	$[m/s^2]$
h_w	=	apparent wall heat transfer coefficient or wall heat transfer coefficient	$[W\cdot m^{-2}\cdot K^{-1}]$
k_l	=	thermal conductivity of liquid	$[W\cdot m^{-1}\cdot K^{-1}]$
k_w	=	wall-to-liquid mass transfer coefficient	[m/s]
L	=	length of electrode in flow direction	[m]
Nu_w	=	Nusselt number for wall-to-liquid heat transfer $(=h_wD/k_l)$	[—]
n	=	exponent in the Richardson-Zaki equation	[—]
ΔP_d	=	pressure drop due to drag force across a particle	[Pa]
ΔP_l	=	dynamic pressure drop	[Pa]
Pr	=	Prandtl number $(=C_{pl}\mu_l/k_l)$	[—]
ΔP_w	=	pressure drop due to wall friction	[Pa]
Sh_w	=	Sherwood number for wall-to-liquid mass transfer $(=k_wD/D_l)$	[—]
Sc	=	Schmidt number $\{=\mu_l/(\rho_lD_l)\}$	[—]
U_g	=	superficial gas velocity	[m/s]
U_i	=	apparent liquid velocity at unit voidage in the Richardson-Zaki equation	[m/s]
U_l	=	superficial liquid velocity	[m/s]
z	=	axial distance	[m]
ε	=	holdup	[—]
μ	=	viscosity	[Pa·s]
ν	=	kinematic viscosity	$[m^2/s]$
ρ	=	density	$[kg/m^3]$

⟨Subscripts⟩

c	=	column wall
g	=	gas
i	=	inner tube

l = liquid
s = solid

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DETERMINATION OF PARTICLE SHAPE FROM ELECTRICAL CONDUCTIVITY OF SUSPENSIONS

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Key Words: Particle, Shape Characterization, Electrical Conductivity, Suspension, Particle Measurement

Experimental investigations are made into the electrical conductivity of suspensions of non-conducting particles of various shapes. Theoretical relations so far available for spheroidal particle systems are tested and the Fricke equation is used for determining particle shape as the aspect ratio of an equivalent spheroid. The electrical conductivities of suspensions of disks and cylindrical particles are obtained experimentally. The effect of the shape distribution on the conductivity of suspensions is also discussed.

Introduction

Particle shape is an important factor that significantly affects the physical characteristics of granular materials. Packed beds of higher density, for example, can be made of particles with higher degree of sphericity and the flowability of particles becomes low for irregular particles.[14] In the surface-finishing process, the rate of grinding metals is enhanced by use of angular abrasive grains.[15] Since the particle shape reflects the process used for their production, one can know the history of the particles from their shape.[7]

The TV image processing system[2,5] accurately measures particle profiles, but it is two-dimensional and usually restricted to a stable orientation, and shape characterization is time-consuming. Moreover, it is in fact probable that the particle shape in only a particular direction is important in shape classification[12,16]

As indirect methods, measurement of the settling velocity leads to an equivalent shape of particles.[3] Gotoh *et al.*[6] related the packing density of particles to the particle shape. The residence time of particles in a system[8,9] may be used as a measure of particle shape, but no theoretical relation has been developed.

In the present study, an experimental investigation is made into the electrical conductivity of suspensions of non-conducting particles of various shapes. First, theoretical relations so far available for spheroidal particle systems are examined and the Fricke equation[4] is chosen to determine the aspect ratio of an equivalent spheroid. The geometrical aspect ratios of non-conducting disks and cylindrical particles are experimentally related to the electrical conductivity of

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