Mass Transfer into a Turbulent Liquid Across the Zero-Shear Gas-Liquid Interface

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Previously published studies have theoretically deduced the mass transfer coefficient k_L on the liquid side by comparing it with the measured values. Most studies have estimated the liquid-side mass transfer coefficient k_L by solving the diffusion equation, together with a two-dimensional, eddy-cell model having the velocity scale V and length scale Λ, and have compared the predicted k_L with the measurements. Then, k_L was given by

$$k_L \sim (D_L V/\Lambda)^{1/2}, \tag{1}$$

where D_L is the molecular diffusivity in liquid. The comparison of Eq. 1 with the measurements suggested which size scales in Eq. 1 were most suitable to explain the measurements of k_L. If the large-scale energy-containing eddies control the mass transfer, V and Λ will be given by the rms value of turbulent velocity fluctuation and integral scale, respectively. For the contribution of small-scale eddies, V and Λ will be given by the Kolmogorov scales, $(\nu/\epsilon)^{1/4}$ and $(\nu^3/\epsilon)^{1/4}$, respectively.

Using this methodology it was deduced that large-scale eddies control the mass transfer in an open-channel flow with grid-generated turbulence (Fortescue and Pearson, 1967), in normal open-channel flows without grids (Komori et al., 1982; Hörner et al., 1986), in an impinging jet, in a thin film flow and in a stirred cell (Davies and Lozano, 1979). On the contrary, Kataoka and Miyauchi (1969) suggested that the mass transfer is controlled by small-scale eddies in an open-channel flow with grid-generated turbulence, and Lamont and Scott (1970) deduced the significant contribution of small-scale eddies by using an eddy cell model. Thus, the discussion on the eddy scales controlling the mass transfer has been controversial and it has not been concluded. This seems to be attributed to the following two reasons.

First, the ratio of the time scale Λ/V of small eddies to that of large eddies is proportional to the square root of the turbulence Reynolds number Re_t, based on the velocity and length scales of large eddies, (Batchelor, 1953); therefore, the relationship between k_L and V/Λ in Eq. 1 is invariable for almost equivalent turbulence Reynolds number flows. This prevents us from determining a contributive eddy size through only the measurements conducted in a turbulent flow with a constant value of Re_t.

The second reason is that the previous works estimated the velocity and length scales by taking conventional averages over all turbulent motions without conditionally averaging surface-renewal motions. The scales have been estimated roughly from the statistical formulae applicable only to ideal isotropic or homogeneous turbulence (e.g., Fortescue and Pearson, 1967). For better estimation of the scales of surface renewal eddies, it is necessary to understand the generation mechanism of renewal eddies and to conditionally average the turbulence signals of renewal eddies by directly detecting renewal motions.

The purpose of this study is to investigate both the physical mechanism of mass transfer across the zero-shear gas-liquid interface and the main contribution of size eddy to mass transfer. To discuss the effects of both generation mechanism of surface-renewal eddies and turbulent Reynolds number Re_t on the mass transfer, two open-channel flows with and without grid-generated turbulence were considered. For a normal open-channel flow without grid-generated turbulence, both the surface-renewal frequency and mass transfer coefficient on the liquid side have been measured by Komori et al. (KMU) (1989), and the same properties were measured only for an open-channel flow with grid-generated turbulence by the same tracer and gas-absorption techniques as in KMU. In addition, they were measured in the same flow field as was done by Fortescue and Pearson (FP) (1967).

Experiments

The same flume as in KMU's open-channel flow without grids was used, and here the turbulence-producing grids were installed at 3.5 m downstream ($x = 0$) of the entrance of the flume perpendicular to the mean flow, as shown in Figure 1a. The grids were of round-rod, square-mesh, single biplane construction, and their mesh size M and the diameter of the rod d were 0.015 to 0.025 m and 0.0009 to 0.005 m, respectively. The cross-sectional mean velocity U_{ave} ranged from 0.05 to 0.14 m/s, and the flow depth δ was maintained at the large values between 0.08 and 0.12 m to avoid the effect of the boundary layer being developed from the flume floor behind grids. The Reynolds number Re_M, based on U_{ave} and M, and the turbulence Reynolds number Re_t, based on the rms value of the streamwise velocity fluctuation measured by the LDV of Komori and Murakami (1988) and integral scale estimated from the power spectrum of streamwise velocity fluctuation, were 810 to 3,490 and 30 to 130 at $x/M = 20$, respectively.

To detect surface-renewal eddies, the same point source technique as in KMU was used. In the open-channel flow with grids, turbulent eddies are generated by each grid; to detect all the eddies, a point source was fixed just above the grid nearest to the free surface, as shown in Figure 1a. Methylene-blue aqueous solution of 0.25 wt.% was emitted from the point source at a speed equal to the ambient-flow velocity. The instantaneous concentration at the free surface in the region behind the grids was measured using the small optical glass-fiber probe of KMU.

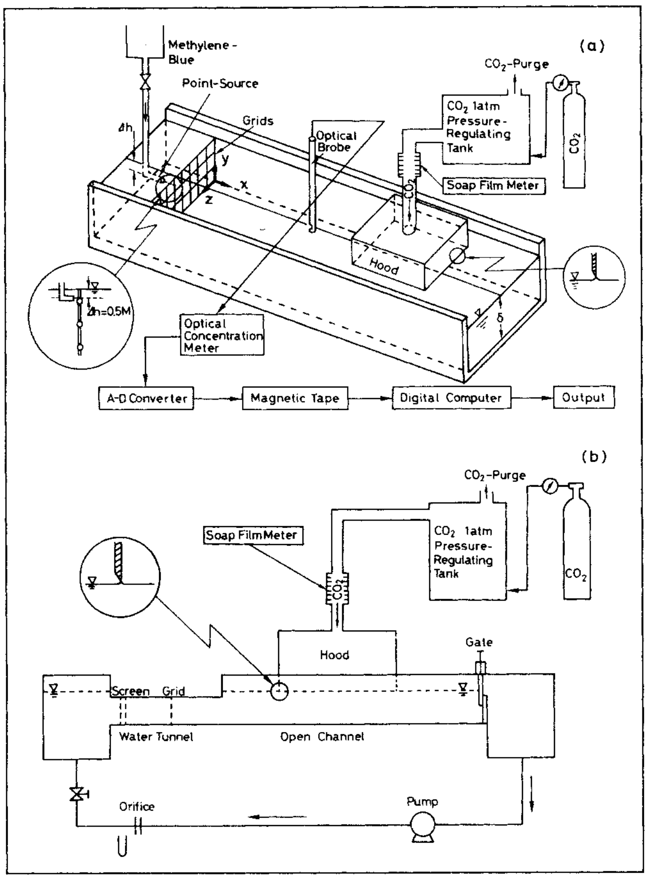

Figure 1. Experimental flumes and instruments.
a. Open channel with turbulence-generation grids
b. Open channel with turbulence-generation grids for the reproduction of the flow of Fortescue and Pearson (1967)

For the additional experiments in the open-channel flow of FP with grids, the same experimental technique was also used in a flume shown in Figure 1b.

Concentration signals were transmitted directly into a digital recorder, and the digital signals were processed by a digital computer. When renewal eddies appear at the free surface, processed concentration signals showed very positively-skewed spikes. To count the number of these spikes, a simple detection function $D_c(t)$ was used;

$$D_c(t) = \begin{cases} 1 \text{ if } c(t) > kc' \\ 0 \text{ otherwise.} \end{cases} \tag{2}$$

The value of the threshold level k was -0.4, which was determined by the same method as in KMU. It was also confirmed by a two-point source technique of KMU that all the surface-renewal eddies arriving at a local measuring point of the free surface are contaminated by the tracer emitted from a point source. Then the frequency f_s of the appearance of renewal eddies can easily be estimated by counting the number N_s of intervals, in which $D_c(t) = 1$ during the measuring period T_{sm} and therefore f_s can be calculated by N_s/T_{sm}. The measured f_s showed a constant value in the region of $x/M > 10$, and its value was adopted here. Furthermore, the scale Λ of the renewal eddy can be estimated approximately from the product of the time interval, in which $D_c(t) = 1$, and the mean surface velocity.

For the estimation of the liquid-side mass transfer coefficient k_L, pure CO_2 at 20°C was absorbed into turbulent water, filling a rectangular hood at atmospheric pressure. The absorption rate of CO_2 was measured by a soap-film meter. From the absorption rate, the liquid-side mass transfer coefficient k_L was estimated. The details of the absorption experiments are given in KMU.

Results and Discussion

Surface renewal frequency and scales of renewal eddies

The correlation between the Reynolds number Re_M and surface-renewal frequency f_s in the open-channel flow with grid-generated turbulence was empirically given by

$$(f_s d/U_{ave})\, Re_M^{0.56} = 0.652\, (f_s\, d^2/\nu)^{0.6}, \tag{3}$$

which was valid for all the turbulence grids used here. The correlation also suggests that surface-renewal motions are generated by rods. For the normal open-channel flow without grid-generated turbulence, the correlation has been given by Eqs. 8 and 9 of KMU, and KMU showed that large-scale renewal motions originate in bursting motions in the buffer layer near the flume floor.

The scales of surface-renewal eddies were estimated from the product of time duration of concentration spikes and mean velocity. The values of the scales were 0.61 ~ 0.90 M for the open-chanel flow with grid-generated turbulence, and the values of 0.37 ~ 1.1 δ were calculated from the data of KMU for the open-channel flow without grids. They were rather scattered among experimental runs, but they showed that the surface renewal eddies have large scales corresponding to the mesh size of grids and the flow depth for the open-channel flows with and without grid-generated turbulence, respectively. The average values of the scales estimated at $x/M = 10$ and $x/\delta = 35$ were 0.76 M and 0.73 δ for the two flows, respectively; and they were close to the integral scales of 0.72 M and 0.70 δ, which were estimated from the power spectra of the streamwise velocity fluctuation.

Surface-renewal eddies vs. mass transfer

The correlation between the mass transfer coefficient k_L and surface-renewal frequency f_s is shown in Figure 2 by using open and solid circles for the open-channel flows with and without grid-generated turbulence, respectively. Here the data for open-channel flows without grids are quoted from KMU. It is found that the correlation is given by the same equation:

$$k_L = 1.4 \times 10^{-5} f_s^{0.5} = 0.34\,(D_L f_s)^{0.5}. \tag{4}$$

The correlation shows that the mass transfer mechanism is invariant between the two turbulent flows with different turbulence production mechanisms and turbulence Reynolds numbers. As mentioned previously, the renewal frequency f_s in Eq. 4 corresponds to the ratio of the velocity scale to the length scale for the large-scale eddies. Therefore, the renewal frequency of the small-scale eddies f_s' may be given by

$$f_s' \sim f_s\, Re_t^{1/2}. \tag{5}$$

This relation is valid generally for grid-generated turbulence (Batchelor, 1956), and it is confirmed also for the free-surface region of a normal open-channel flow without grids by Nakagawa et al. (1975). If small-scale eddies control the mass transfer across the interface, k_L will be correlated with f_s':

$$k_L \sim (D_L f_s')^{1/2}. \tag{6}$$

Substituting Eq. 5 into Eq. 6, we obtain

$$k_L \sim (D_L f_s)^{1/2}\, Re_t^{1/4}. \tag{7}$$

This equation suggests that, beside f_s, $Re_t^{1/4}$ should strongly affect k_L for the small eddy control. However, when k_L is plotted against $f_s^{1/2}\, Re_t^{1/4}$, k_L is not proportional to $f_s^{1/2}\, Re_t^{1/4}$, as shown

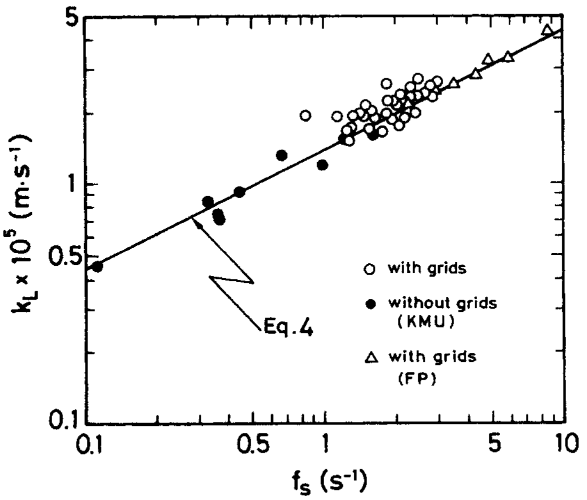

Figure 2. Correlation between surface-renewal frequency and liquid-side mass transfer coefficient.

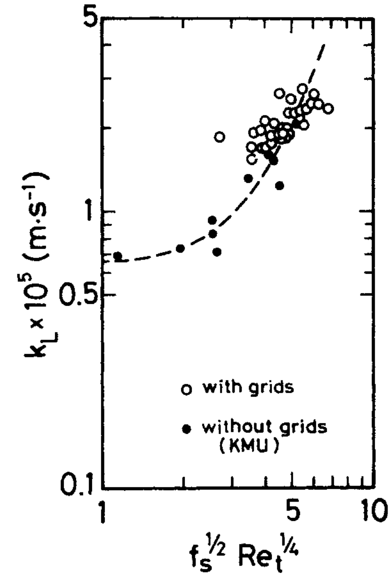

Figure 3. Correlation between $f_s^{1/2}\, Re_t^{1/4}$ and liquid-side mass transfer coefficient.

in Figure 3. Here the values of $Re_t^{1/4}$ were estimated at $x/M = 20$, and the values of Re_t in the KMU's open-channel flow without grids were 100 to 420. This shows that the mass transfer should not be controlled by small-scale eddies, but by large-scale eddies.

It should be noted that the real dimensions of large-scale eddies are quite different in the present two flows and that the term "large scale" means the scale comparable to the integral scale. Thus, such a large-scale eddy cell model, as proposed by FP, is expected to be most suitable to explain mass transfer across gas-liquid interfaces. The constant of 0.34 in Eq. 4 is, however, rather smaller than the value close to the unity used in a large-eddy cell model of FP. This difference is not caused by the difference of the mass transfer mechanism or the water quality, but it is attributed to the fact that the flow of FP is not an ideal, grid-generated turbulence, but a complicated flow mixed with both grid-generated turbulence and wall turbulence in an open channel. That is, the scales of renewal eddies, which were estimated by FP on the assumption of ideal grid-generated turbulence, may contain noticeable errors; and the agreement between the measured and predicted values of k_L in FP may be a mere accidental result.

In fact, the surface-renewal frequency, f_s, measured in the FP's flow reproduced in the flume of Figure 1b by using the same point-source technique, was somewhat larger than the f_s of Eq. 3 measured in the present open-channel flow with grid-generated turbulence. This clearly showed that in the flow of FP large-scale renewal eddies are generated by bursting, in addition to grid-generated renewal eddies. When f_s is also plotted against k_L measured in the reproduced flow of FP, k_L is well correlated by Eq. 4, as shown by triangles in Figure 2. This shows that the mass transfer mechanism is quite the same between the present open-channel flows and the flow of FP, in which large-scale eddies control mass transfer across the interface. The original experiments of FP can be confirmed by plotting k_L against Re, as shown in Figure 4. The values of k_L measured in the reproduced flow of FP are in good agreement with the empirical

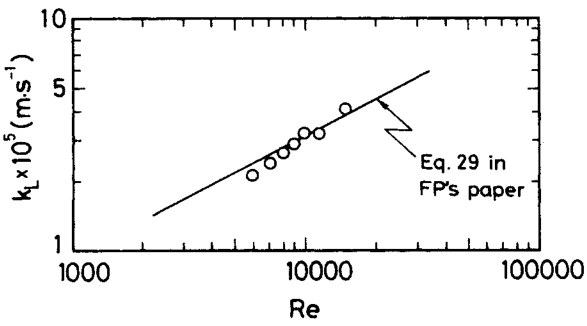

Figure 4. Variation of k_L against the Reynolds number in Fortescue and Pearson's (1967) flow.

formula (Eq. 29) given by FP. This confirms that the experiments of FP are very carefully reproduced here and that the above discussion of FP's experiments are quite rational.

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Notation

c = concentration fluctuation of tracer
c' = rms value of c
d = diameter of rod
$D_c(t)$ = detection function in Eq. 2
D_L = molecular diffusivity in liquid
f_s = surface-renewal frequency by large eddies
k_L = mass transfer coefficient on the liquid side
M = mesh size of grid
Re_M = Reynolds number based on the mesh size M and cross-sectional mean velocity U_{ave}
Re_t = turbulence Reynolds number based on the rms value of streamwise velocity fluctuation and integral scale
U_{ave} = cross-sectional mean velocity
V = velocity scale of renewal eddy
x = streamwise co-ordinate

Greek letters

δ = flow depth
Λ = length scale of renewal eddy
ϵ = viscous dissipation
ν = kinematic viscosity

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