SIMULATING AIR-WATER GAS TRANSFER AT HIGH SCHMIDT NUMBER USING DYNAMICS OF OPEN-CHANNEL TURBULENCE

Yuji SUGIHARA¹, Daisuke NAKAGAWA², Ryosuke TERAOKA³ and Koji SHIONO⁴

¹Member of JSCE, Assoc. Professor, Dept. of Earth System Science and Technology, Kyushu University
(6-1 Kasuga-Koen, Kasuga 816-8580, Japan)
E-mail: sugihara@esst.kyushu-u.ac.jp
²Mitsubishi Hitachi Power Systems Co., LTD. (6-9 Takaramachi, Kure 737-8508, Japan)
E-mail: daisuke1_nakagawa@mhps.com
³Student Member of JSCE, Graduate Student, Dept. of Earth System Science and Technology, Kyushu University
(6-1 Kasuga-Koen, Kasuga 816-8580, Japan)
E-mail: teraoka@esst.kyushu-u.ac.jp
⁴Professor, Dept. of Civil and Building Engineering, Loughborough University
(Loughborough, Leicestershire, LE11 3TU, UK)
E-mail: K.Shiono@lboro.ac.uk

We propose a simple numerical method to describe the air-water gas transfer at high Schmidt number. A numerical simulation is carried out in terms of the advection-diffusion equation using interfacial velocities obtained from a direct numerical simulation of open-channel turbulent flow. The present numerical method can describe the high Schmidt number air-water gas transfer driven by turbulence dynamics of the open channel flow. The numerical results show that the surface divergence effectively promotes the interfacial gas flux. Characteristics of surface-renewal eddies, such as surface divergence and vorticities, are found on the basis of cross-correlation coefficients between the interfacial gas flux and turbulent quantities.

Key Words: gas transfer, open channel, air-water interface, surface divergence and Schmidt number

1. INTRODUCTION

Clarification of the air-water gas transfer mechanism for carbon dioxide and oxygen is important for developing techniques to evaluate quantitatively the biological and water-quality environments in rivers and seas. Problems in interfacial scalar transport have attracted attention as fundamental studies of gas exchange across the air-sea interface, which is concerned with the prospect of global warming. For lower-dissolved gas such as carbon dioxide and oxygen, the concentration boundary layer on the liquid side is much thinner than that on the air side, and the interfacial scalar transport becomes the liquid-side resistance, so that the gas flux at the air-water interface is controlled by turbulence on the liquid side. Therefore, the main problem in gas transfer studies on hydraulic engineering is to establish a method to estimate quantitatively the gas transfer velocity using turbulent characteristic quantities on the liquid side.

Previous studies on the gas transfer velocity at the air-water interface had tried to investigate whether turbulent eddies to control the gas transfer were large eddies at the energy-containing range or smaller eddies at the energy-dissipative range¹,²,³. Chu and Jirka⁴ measured the dissolved oxygen and the structure of turbulence close to the air-water interface in grid-stirred turbulence by means of micro oxygen-electrode and hot-film velocimetry. They pointed out that the gas transfer in such a turbulent field may be described by large eddies’ scaling. On the other hand, using similar turbulent systems, Asher and Pankow⁵ showed the turbulent eddies controlling the gas transfer to be smaller ones. Several experimental studies have been conducted on the gas transfer velocity in open-channel flows. Moog and Jirka⁶ carried out laboratory measurements in open-channel flows at high Reynolds number and reported that the gas transfer velocity can be described by the small-eddy model.

Besides the large- and small-eddy conceptual models, some researchers have also examined the gas transfer mechanism, focusing on the divergence of horizontal velocities at the air-water interface, i.e., the surface divergence. McCready et al.⁷ proposed a model that associated the gas transfer velocity with the root-mean-square of the surface divergence on the
basis of an unsteady advection-diffusion equation of dissolved gas. Due to recent developments in measuring techniques, direct measurements of the surface divergence have been made by using the techniques of PIV\(^8\). Tsumori and Sugiura\(^9\) measured the turbulent structure on the air-water interface in grid-stirred turbulence by means of PIV, and they showed that the gas transfer velocity can be expressed by the root-mean-square of the surface divergence. Also, Sanjou and Nezu\(^10\) carried out detailed measurements in open-channel flows using the PIV techniques. They found out that a quantitative relationship between the gas transfer velocity and the surface divergence in open-channel flow was different from that in grid-stirred turbulence. They then proposed a modified surface divergence model.

As mentioned above, various data to quantify the gas transfer have been collected from the standpoint of experimental studies. However, there may be limitations in understanding the unsteady and microscopic turbulent phenomena by experimental approaches only. Remarkable developments in computers allow us to investigate the gas transfer through direct numerical simulations (DNS) for turbulence in open-channel flows\(^{11,12,13}\). However, DNS can be made under conditions at low Schmidt number only, because the thickness of dissolved gas on the liquid side becomes in the order of \(1/\sqrt{ReSc}\) relative to macro turbulent scale. Schmidt number \(Sc\) is defined as a dimensionless number expressing the ratio of momentum transfer to molecular dissolved-gas diffusion, and it seems to be difficult to carry out DNS at high Schmidt number even to the latest computer environments. As one of numerical strategies for the gas transfer at high Schmidt number, Hasegawa and Kasagi\(^14\) proposed a hybrid scheme of DNS and LES, and they made a numerical simulation at high Schmidt number.

The present study is intended to propose a simple method to simulate the gas transfer at high Schmidt number by analyzing the advection-diffusion equation of dissolved gas using velocity data on the air-water interface obtained from DNS; thus, the numerical simulation of the gas transfer can be done only within the concentration boundary layer very close to the air-water interface. Our analytical object is an open-channel flow, and we can reproduce the gas transfer mechanism at the air-water interface driven by the dynamics of open-channel turbulence through this simple method.

### 2. ANALYTICAL MODEL OF GAS TRANSFER AT HIGH SCHMIDT NUMBER

This study aims at understanding gas transfer in open-channel flow. The governing equations are normalized by using the depth of open-channel \(H\), the wall friction velocity \(u_r\), the dissolved gas concentrations on the air-water interface \(c_s\) and in the bulk region \(c_b\). The velocities, the concentration, the time and the spatial coordinates are defined as follows:

\[
\begin{align*}
\bar{u}_s &= \frac{u}{u_r}, \quad \bar{v}_s = \frac{v}{u_r}, \quad \bar{w}_s = \frac{w}{u_r}, \quad c_s = \frac{c - c_b}{c_s - c_b} \\
\bar{x}_s &= \frac{x}{H}, \quad \bar{y}_s = \frac{y}{H}, \quad \bar{z}_s = \frac{z}{H}, \quad \bar{t} = \frac{t}{u_r/H}
\end{align*}
\]

where * denotes the dimensionless quantity. The coordinate system is shown in Fig.1, where \(x_s, y_s\) and \(z_s\) are the streamwise, spanwise and vertical-downward directions, respectively. We consider the concentration boundary layer to be much thinner compared to surface renewal eddies’ scale; thus, the vertical variations of the horizontal velocities and the horizontal molecular diffusion can be neglected in the present analysis.

First, we obtain the following equation from the equation of continuity:

\[
\frac{\partial \bar{w}_s}{\partial \bar{z}_s} = -\left(\frac{\partial \bar{u}_s}{\partial \bar{x}_s} + \frac{\partial \bar{v}_s}{\partial \bar{y}_s}\right) = -\beta_s(x_s, y_s, \bar{t})
\]

where \(\bar{u}_s, \bar{v}_s\) are the horizontal velocities and \(\beta_s\) indicates the surface velocity divergences, i.e., the surface divergence. Assuming that the fluctuation of the interface is very small and \(\bar{w}_s = 0\) at \(\bar{z}_s = 0\), we can express the surface divergence by analyzing the advection-di

### Table 1 Numerical conditions.

<table>
<thead>
<tr>
<th>Spatial derivative</th>
<th>4th-order central difference scheme (excepting at vertical boundary)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time evolution</td>
<td>2nd-order Adams-Bashforth method</td>
</tr>
<tr>
<td>Size of computational domain</td>
<td>Streamwise : (x) = 7.68</td>
</tr>
<tr>
<td></td>
<td>Spanwise : (y) = 3.84</td>
</tr>
<tr>
<td></td>
<td>Vertical : (z) = 0.024</td>
</tr>
<tr>
<td>Number of grid</td>
<td>((N_x, N_y, N_z) = (640, 480, 81))</td>
</tr>
<tr>
<td>Grid spacing</td>
<td>((\Delta x, \Delta y, \Delta z) = (1.2 \times 10^{-2}, 8.0 \times 10^{-2}, 1.5 \times 10^{-2}))</td>
</tr>
<tr>
<td>Time difference</td>
<td>(\Delta t = 5.0 \times 10^{-3})</td>
</tr>
<tr>
<td>Total computation time</td>
<td>(20000) steps ((t = 1.0))</td>
</tr>
<tr>
<td>Dimensionless parameters</td>
<td>(Re_{\tau} = 150, Sc = 600)</td>
</tr>
<tr>
<td>Boundary conditions</td>
<td>(x) - and (y) - boundaries Periodic</td>
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<td></td>
<td>(z) - boundary</td>
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<tr>
<td></td>
<td>(c = 1) at (z = 0)</td>
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<tr>
<td></td>
<td>(\partial c/\partial z = 0) at (z = 0.024)</td>
</tr>
</tbody>
</table>
the vertical velocity \( w_s \) as follows:
\[
w_s = -\beta_s(x_s, y_s, t_s)z_s^2
\]
(3)

From the above approach, the dimensionless advection-diffusion equation of dissolved gas is given by
\[
\frac{\partial c_s}{\partial t_s} + (U_x + u'_x) \frac{\partial c_s}{\partial x_s} + v'_z \frac{\partial c_s}{\partial z_s} - \beta_s z_s \frac{\partial c_s}{\partial z_s} = \frac{1}{Re_s Sc} \frac{\partial^2 c_s}{\partial z_s^2}
\]
(4)

\( U_s \) in the equation is the streamwise velocity averaged over the air-water interface, \( u'_x \) and \( v'_z \) the velocity fluctuations. For the analysis in the present study, in order to observe easily the gas transport phenomena, the governing equations are transformed from the fixed coordinate system to a moving system at the mean streamwise velocity. By defining the coordinate system moving at the mean streamwise velocity as \( \hat{x}_s = x_s - U_s t_s \), Eq.(4) becomes the following equation:
\[
\frac{\partial c_s}{\partial t_s} + (\hat{U}_x + u'_x) \frac{\partial c_s}{\partial \hat{x}_s} + v'_z \frac{\partial c_s}{\partial z_s} - \beta_s z_s \frac{\partial c_s}{\partial z_s} = \frac{1}{Re_s Sc} \frac{\partial^2 c_s}{\partial z_s^2}
\]
(5)

Hereafter, \( \hat{x}_s \) is described as \( x_s \) for brevity.

For the horizontal velocities in Eq.(5), we use the interfacial velocities obtained from DNS in open-channel flows by Sugihara et al.\(^{(13)}\) Their DNS code was based on Kajishima’s opened code for the incompressible Navier-Stokes equation with the Simplified MAC method. In this study, the velocity data are interpolated using the spline interpolation to make much finer numerical grids than those of DNS. The boundary conditions of dissolved gas are imposed such that \( c_s = 1 \) at \( z_s = 0 \) and the vertical concentration gradient becomes zero at the lower boundary of the concentration boundary layer. The computation is also carried out under the initial conditions that \( c_s = 1 \) at \( z_s = 0 \) and \( c_s = 0 \) anywhere except at \( z_s = 0 \).

Dimensionless parameters to describe the phenomena are Reynolds number \( Re_s(=u_L H/\nu) \) and Schmidt number \( Sc(=\nu/D) \), where \( \nu \) is the kinematic viscosity of fluid on the liquid side and \( D \) is the molecular diffusivity of dissolved gas. By assuming carbon dioxide as dissolved gas, \( Re_s \) and \( Sc \) become 150 and 600, respectively. The computational region in the vertical direction ranges from the interface to 2.4% of the open-channel depth, and the vertical grid points of 81 are put in a non-uniform grid system so as to be fine near the interface. The computations are carried out for 20,000 steps, corresponding to the unit dimensionless time with a dimensionless time difference of \( 5.0 \times 10^{-5} \). For the time integration of the governing equations, the Eulerian explicit method is used for the first step, whereas after the second step, the second-order Adams-Bashforth method is applied. The details of the numerical conditions are summarized in Table 1.

3. ANALYTICAL RESULTS AND DISCUSSION

(1) Relationship between interfacial turbulence and interfacial gas flux

Figure 2 shows the spatial distributions of the surface divergence \( \beta_s \) and the interfacial gas flux \( F_s \), where (a) and (b) indicate the surface divergence and the interfacial gas flux, respectively. The interfacial gas flux is defined by
\[
F_s = -\frac{1}{Re_s Sc} \frac{\partial c_s}{\partial z_s}
\]
(6)

The upper and lower panels are the spatial distributions on the interface \( (z_s=0) \) at \( t_s = 0.05 \) and 0.75, respectively. In these figures, the regions of the larger values of the surface divergence and the downward flux are shown by the red-colored contours. It can be recognized that the downward flux at \( t_s = 0.05 \) takes large values over the whole region. This is because the vertical gradient of the concentration at the interface becomes large anywhere at the initial stage of the computation. As time elapses, such a pattern is moderated, and it can be seen from the results at \( t_s = 0.75 \) that the region of the positive surface divergence agrees obviously with that of the large downward flux. This means that the surface divergence has an important role in the gas transfer at the air-water interface. In the region of large surface divergence, the upward current with lower-concentration fluid induces the surface divergent motion. The divergent motion stretches the concentration boundary layer horizontally, and the interfacial gas flux increases due to the increase of the vertical concentration gradient.

Figures 3(a) and 3(b) show spatial distributions on the interface for the vorticity in the streamwise direction \( \omega_{sz} \) and that in the spanwise direction \( \omega_{sz} \), respectively. It should be noted that vertical velocities are needed for calculations of these vorticities, which are done using the vertical velocities at the depth of 1 grid point beneath the interface \( (z_s = 1.5 \times 10^{-3}) \). The figures show that turbulent eddies with the rotational axis parallel to the interface are set densely in the regions where the interfacial gas flux and the surface divergence take large values. The distributions of these vorticities may be recognized as the spatial visualization pattern of each directional component of the horizontal vorticity-vectors due to surface-renewal eddies. We guess that the dynamics of these eddies has an important role in generating the strong positive surface divergence. Because of free-sheared interference, the surface-renewal eddies may be originated from the lift-up of turbulent eddies generated by the bursting of longitudinal vortices near the bottom wall. Statistical characteristics of the surface-renewal eddies will be discussed later.

Figures 4 and 5 show the time evolutions of the
vertical profiles of the mean gas concentration and the root-mean-square (rms) of the gas concentration fluctuation, which are obtained from the spatial averaging in each horizontal plane parallel to the interface. In the figures, the vertical profiles are plotted for six cases of $t_s=0.01, 0.05, 0.10, 0.25, 0.50$ and $0.75$. The results show that the mean concentration decreases monotonously with the depth, but the rms of the fluctuation takes a maximum value near the interface. Focusing on the time variation, we can observe that the boundary thickness of the mean concentration increases monotonously as time elapses. The value of the rms becomes large over the whole computation region, and the depth for the maximum rms-value increases with time.

We attempt to calculate the boundary layer thickness of the mean concentration through the following analytical solution. The advection-diffusion equation shown in Eq.(5) is simplified by neglecting the horizontal advection terms and using the surface divergence as constant. Under the boundary condition that $c_s \to 0$ as $z_s \to \infty$, the analytical solution for the simplified equation becomes

$$\frac{c_s - c_{bs}}{c_{ss} - c_{bs}} = 1 - \frac{2}{\sqrt{\pi}} \int_0^\infty \exp(-\zeta^2) d\zeta \quad \zeta = \frac{z_s}{\delta_s(t_s)} \quad (7)$$

In this study, the thickness of the concentration boundary layer is calculated by fitting the analytical solution of Eq.(7) into the vertical profile of the mean concentration from the numerical simulation. Figure 6 shows the time evolutions of the spatially-averaged values of the concentration boundary layer thickness $\delta_s$ and the interfacial gas flux $F_s$. This figure shows that the concentration boundary layer thickness increases rapidly at the initial stage of the computation, but its temporal variation becomes milder as time
(2) Statistical characteristics of interfacial turbulence controlling gas transfer

Statistical characteristics of the surface renewal eddies that induce the interfacial gas flux are investigated on the basis of the cross-correlation coefficients between the interfacial gas flux and turbulent characteristic quantities. The cross-correlation coefficients are defined as the following relations, and each relation represents the cross-correlation coefficient in the \( x \) or \( y \) direction:

\[
C_x(r_1) = \frac{\left< F'(x_s, y_s) A'(x_s + r_1, y_s) \right>}{F_{\text{rms}} A_{\text{rms}}} \tag{8}
\]

\[
C_y(r_2) = \frac{\left< F'(x_s, y_s) A'(x_s, y_s + r_2) \right>}{F_{\text{rms}} A_{\text{rms}}} \tag{9}
\]

where \( F'(x_s, y_s) \) is the spatial fluctuation component of the interfacial gas flux at \( (x_s, y_s) \). \( A' \) denotes turbulent quantities, which correspond to the spatial fluctuation components of the surface divergence, the streamwise and spanwise vorticities in this study. It is noticed that the suffix of \( \text{rms} \) in the equations indicates the root-mean-squared quantity, and \( < > \) means temporally and spatially averaged one.

Figures 7(a) and 7(b) show the cross-correlation coefficients between the interfacial gas flux and the turbulent quantities from Eqs. (8) and (9), respectively. It is seen from the coefficients in both directions that the interfacial flux is most strongly correlated with the surface divergence. In addition, for the vorticity in the direction normal to the distance \( r \), the coefficients show inverse correlation across \( r=0 \). This suggests that the interfacial flux can be promoted effectively in the case where turbulent eddies with the rotation axis parallel to the interface are arranged so as to induce the upwelling flow of low-concentration fluid, i.e., the positive surface divergence, at a target point. Handler et al.\(^{11}\) and Nagaosa and Handler\(^{12}\) carried out the direct numerical simulations under the conditions that the Prandtl number is 2 and 1, respectively, and analyzed the cross-correlation coefficients in a similar way. Our results at high Schmidt number confirmed roughly the same characteristics as the ones for the spanwise cross-correlation coefficients. However, among the streamwise cross-correlation coefficients, our results for the coefficient with the spanwise vorticity are quite different from characteristics of their results. For the case at high Schmidt number, the symmetry of the amplitudes of both correlation coefficients across \( r_1=0 \) is remarkable, while at low Prandtl number, which is equivalent to the case at low Schmidt number, the asymmetry of the amplitudes becomes larger. The physical reasons why such a difference occurs are not clear at this stage, but this suggests that the structure of surface renewal eddies promoting the interfacial flux is different between at high Schmidt number and low high Schmidt number. This finding is of interest from the viewpoint of interfacial scalar transport, and we will examine this point in detail in the future.
We compare the present numerical results with a local gas transfer model effective for the vicinity of the flow stagnation where the surface divergence dominates, assuming an ideal situation when the fluctuation of the interface can be ignored. Supposing a local equilibrium state at which the vertical advection term expressed in the surface divergence is balanced by the vertical diffusion term, the advection-diffusion equation to be reduced is given as follows:

\[ \frac{\partial c}{\partial t} + \beta \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{\partial z^2} \]  

(10)

where the temporal variation in the surface divergence is assumed to be sufficiently small and the divergence remains constant. This model equation becomes similar as that examined by Awaya and Abe\(^{(15)}\). Our analytical target is only for the regions of the positive surface divergence, i.e., \( \beta > 0 \). In addition, the boundary conditions are given by

\[
\begin{align*}
  c &= c_s & \text{at} & z = 0 \\
  c &= c_b & \text{as} & z \to \infty 
\end{align*}

(11)

Solving Eq.(10) under the boundary conditions of Eq.(11), we obtain the following analytical solution:

\[
\frac{c - c_b}{c_s - c_b} = 1 - \frac{2}{\sqrt{\pi}} \int_0^\eta \exp(-\eta^2) d\eta
\]

(12)

From Eq.(12), we obtain the local surface flux \( F_c \) in the regions of the positive surface divergence as follows:

\[
F_c = -D \frac{dc}{dz} = \sqrt{\frac{2}{\pi}} \sqrt{\Delta \beta} (c_s - c_b)
\]

(13)

The interfacial gas flux can be expressed in the form of the product of the gas transfer velocity with the concentration difference between the interface and the bulk region, thus the local gas transfer velocity can be given by

\[
k_L = \sqrt{\frac{2}{\pi}} \sqrt{\Delta \beta}
\]

(14)

From the viscous wall unit scaling with the friction velocity \( u_* \) and the kinematic viscosity \( \nu \), the dimensionless gas transfer velocity is obtained as follows:

\[
k_L^+ = \sqrt{\frac{\nu}{u_*}} Sc^{-1/2} \sqrt{\beta^+}
\]

(15)

where \( + \) is the dimensionless quantity expressed in the viscous wall unit. In order to verify the validity of the local gas transfer model, Fig.8 shows the comparison of Eq.(15) with the numerical results from the interfacial flux, which are filtered by the moving average in an infinitesimal interval \( \Delta \beta^+ \). The \( \Delta \beta^+ \) is 0.01 in the range from \( \beta^+ = 0 \) to 0.1, and 0.02 in the other range because of the small number of data. In this figure, the behavior of Eq.(15) is also drawn by the red line to compare the numerical results with the gas transfer model. Equation (15) only stands up in the positive surface divergence region, but the numerical results do not have such a limitation. The disparity between both for the surface divergence from the vicinity of 0 to the negative region reflects this precondition. This figure shows that the local gas transfer velocity for the negative divergence takes lower values and does not change much, whereas that for the positive divergence increases with the local surface divergence. Furthermore, the behavior in the positive surface divergence region shows good agreement with Eq.(15). This indicates that the local gas transfer velocity increases in proportion to the root of the local surface divergence, and the gas transfer in the negative divergence region may be ruled by a mechanism different from the surface divergence model.

4. CONCLUSIONS

In this study, we proposed the analytical model to describe the air-water gas transfer at high Schmidt number, for which the numerical calculation was made only within the concentration boundary layer by

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