Gas Absorption with First-Order Chemical Reaction into a Turbulent Falling Liquid Film

M.R. Riazi[1] and J. Razavi[2]

A numerical solution of the governing partial differential equation for gas absorption in turbulent falling liquid films with and without the first order homogeneous reaction and external gas phase mass transfer is shown. The best eddy diffusivity model to describe the flow distribution is the van Driest model, modified in the outer region of the film by the use of an eddy diffusivity deduced from gas absorption measurements. The results are given for special cases to illustrate the effect of turbulence on concentration profile and the rates of gas absorption.

INTRODUCTION

From the review of published literature on wetted-wall columns, it is evident that, while the problem of gas absorption with or without chemical reaction in laminar falling liquid films has been studied in some detail, the case of turbulent flow, especially when chemical reaction is involved, has received less attention. The problem of gas absorption without chemical reaction in laminar films was solved by Olbricht and Wild [1] using the series expansion method. For the case of first-order chemical reaction in laminar films, many investigators solved the governing equation by the method of separation of variables [2,3] or numerical techniques [4]. The problem of physical gas absorption into a turbulent liquid film was treated to some extent by Lamourelle and Sandall [5]. Based on the gas absorption measurements, they obtained an expression for the liquid phase eddy diffusivity in the region near the free surface. Menez

and Sandall [6] studied the problem of gas absorption accompanied by first-order chemical reaction in a liquid flowing in fully developed turbulent flow. They obtained asymptotic solutions for which solute concentrates only a short distance into the liquid film because of a slow rate of diffusion or very high rate of reaction where only eddy diffusivity in the region near the free surface was used to describe the turbulence in the liquid film.

For our work, the eddy diffusivity given by Lamourelle and Sandall [5] is used for the region near the free surface, while the van Driest viscosity model has been used for the region near the wall.

FORMULATION

Let us consider the system shown in Figure 1. A liquid initially free of the absorbing spieces at $z = 0$ flows down the surface of a vertical and impermeable wall under the influence of gravity.

1. Department of Chemical Engineering, College of Engineering and Petroleum, Kuwait University, P.O. Box 5969, Safat 13060, Kuwait.
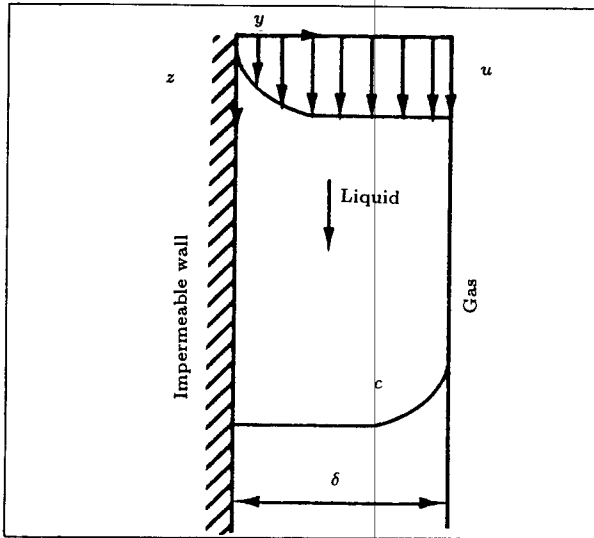2. Department of Chemical Engineering, Sharif University of Technology, Tehran, I.R. Iran.

Figure 1. Schematic of a falling liquid film showing the coordinate system.

The absorbings spieces are absorbed by the liquid where it undergoes a (pseudo) first-order irreversible chemical reaction. It is assumed that the gas phase concentration of absorbing spieces is constant and the interfacial shear stress at the gas-liquid interface is neglected. Furthermore, it is assumed that the diffusion in the axial direction is negligible. Under these conditions, the steady state mass balance on the absorbing spieces in the liquid phase for turbulent flow is:

$$u\frac{\partial C}{\partial z} = \frac{\partial}{\partial y}(D + \varepsilon_D)\frac{\partial C}{\partial y} - kC \ . \tag{1}$$

The coordinate system used and the physical description of the absorbing film are shown in Figure 1. u is the axial velocity of liquid film and can be found from the momentum equation after neglecting the pressure gradient and axial terms:

$$\frac{\partial}{\partial y}[(\nu + \varepsilon_M)\frac{\partial u}{\partial y}] + g = 0 \ . \tag{2}$$

The solution of the above equations for momentum and mass transfer requires the specifications of the boundary and initial conditions:
at inlet $z = 0$,

$$C = 0 \ , \tag{3}$$

at wall $y = 0$,

$$u = 0 \ ,$$

$$\frac{\partial C}{\partial y} = 0 \ , \tag{4}$$

at interface $y = \delta$,

$$\frac{\partial u}{\partial y} = 0 \ ,$$

$$\frac{\partial C}{\partial y} = \frac{k_G}{D}(C^* - C) \ , \tag{5}$$

where δ is the film thickness, C^* is the concentration in equilibrium with gas phase and k_G is the mass transfer coefficient in the gas phase. Upon integration of Equation 2, with corresponding boundary conditions given in Equations 4 and 5, the velocity profile can be obtained:

$$u = \int_0^y \frac{g(\delta - y)}{\nu + \varepsilon_M}dy \ . \tag{6}$$

In the above equations, if flow is laminar, $\varepsilon_M = \varepsilon_D = 0$.

Introducing the following nondimensional variables: $\overline{y} = \frac{y\sqrt{g\delta}}{\nu}$, $\overline{u} = \frac{u}{\sqrt{g\delta}}$, $\overline{z} = \frac{z\sqrt{g\delta}}{\nu}$, $\overline{\delta} = \frac{\delta\sqrt{g\delta}}{\nu}$, $\overline{C} = \frac{C}{C^*}$, will transform Equations 1, 3, 4, 5 and 6 to the following form:

$$\overline{u}\frac{\partial \overline{C}}{\partial \overline{z}} = \frac{\partial}{\partial \overline{y}}(\frac{1}{Sc_t} + \frac{1}{Sc_t}\frac{\varepsilon_D}{\nu})\frac{\partial \overline{C}}{\partial \overline{y}} - \frac{\alpha}{\overline{\delta}^{\frac{2}{3}}}\overline{C} \ , \tag{7}$$

at $\overline{z} = 0$ and $0 \leq \overline{y} \leq \overline{\delta}$,

$$\overline{C} = 0 \ , \tag{8}$$

at $\overline{y} = 0$,

$$\frac{\partial \overline{C}}{\partial \overline{y}} = 0 \ , \tag{9}$$

at $\overline{y} = \overline{\delta}$,

$$\frac{\partial \overline{C}}{\partial \overline{y}} = \frac{N}{\overline{\delta}^{\frac{1}{3}}}(1 - \overline{C}) \ , \tag{10}$$

and

$$\overline{u} = \int_0^{\overline{y}} \frac{(1 - \frac{\overline{y}}{\overline{\delta}})}{\overline{\varepsilon}_M}d\overline{y} \ , \tag{11}$$

where $\alpha = k(\frac{\nu}{g^2})^{\frac{1}{3}}$, $N = \frac{k_G}{D}(\frac{\nu^2}{g})^{\frac{1}{3}}$ and $\overline{\varepsilon}_M = 1 + \frac{\varepsilon_M}{\nu}$.

Note that when the mass transfer resistance in the gas phase is negligible, $N = \infty$, the boundary condition given in Equation 10, reduces to $\overline{C} = 1$ at $\overline{y} = \overline{\delta}$.

In order to proceed to solve the equations for the turbulent case, it is necessary to introduce some empirical profiles for the eddy diffusivity. Some typical models for the falling film are introduced by Seban and Fari [7]. Accurate specifications of the eddy diffusivity close to the wall, and also close to the free surface, are much more important than in the middle of the film, due to low resistances in the central region. It is customary that, for modelling ε_M, the flow is divided into two regions, an inner region where the turbulent transport is dominated by the presence of the wall and an outer wall-like region. The best model to describe the prediction of evaporation, heating and gas absorption is the van Driest model, modified in the outer region of the film by use of an eddy diffusivity deduced by Lamourelle and Sandall [5] from gas absorption measurement [7].

Various assumptions have been made in order to describe the mean velocity distribution near the wall. A popular kinematic eddy viscosity model for this region, as mentioned before, is provided by an expression by van Driest [8], who assumed the following modified expression for the Prandtl mixing length theory:

$$L' = K'y[1 - exp(\frac{y}{A})] ,$$

where A is a damping length constant defined as $26\nu\sqrt{\frac{\rho}{\tau_o}}$ and $K' = 0.4$. For the falling film, van Driest viscosity is used in the inner layer of film liquid. Therefore, for the inner region:

$$\varepsilon_M = L'^2|\frac{du}{dy}| ,$$

$$(\nu + L'^2|\frac{du}{dy}|)\frac{du}{dy} = g(\delta - y) ,$$

$$|\frac{du}{dy}| = \frac{-\nu + \sqrt{\nu^2 + 4L'^2 + 4L'^2(\delta - y)g}}{2L'^2} ,$$

$$\varepsilon_M = \frac{-\nu + \sqrt{\nu^2 + 4L'^2 + 4L'^2(\delta - y)g}}{2} .$$

Nondimensionalization transfers the above equation in the following form:

$$\overline{\varepsilon}_M = \frac{1+\sqrt{1+0.64\overline{y}^2[1-exp(-\frac{\overline{y}}{26})]^2(1-\frac{\overline{y}}{\delta})}}{2} . \quad (12)$$

Lamourelle and Sandall [5], by measuring the mass-transfer coefficient for the liquid phase for gas absorption into a turbulent liquid flow down a wetted column, obtained the following pattern distribution for ε_D at a temperature of $25°C$:

$$\varepsilon_D = 0.284 Re^{1.678}(\delta - y)^2 , \quad (13)$$

in which ε_D is in $\frac{ft^2}{hr}$. In order to generalize the above equation to temperatures other than $25°C$ and to liquid other than water, it must be rendered dimensionless in a manner which Levich [9] has indicated:

$$\frac{\varepsilon_D}{\nu} \propto k''\frac{g\rho}{g_c\sigma} ,$$

where k'' is a constant and σ is the surface tension. The above constant can be calculated from Equation 13, which yields the following result:

$$\frac{\varepsilon_D}{\nu} = 6.4 \times 10^{-4}\frac{g\rho}{g_c\sigma}Re^{1.678}(\delta - y)^2 ,$$

which, for computation purposes upon nondimensionalization, reduces to:

$$\overline{\varepsilon}_D = 1 + 6.47 \times 10^{-4}\frac{g^{\frac{1}{3}}\rho\nu^{\frac{3}{4}}}{g_c\sigma\overline{\delta}^{\frac{2}{3}}}Re^{1.678}(\overline{\delta} - \overline{y})^2 . \quad (14)$$

The Reynolds number is defined as:

$$Re = \frac{4\Gamma}{\mu} = \frac{4\int_0^\delta \rho u dy}{\mu} . \quad (15)$$

In dimensionless form, Equation 15 becomes:

$$Re = 4\int_0^{\overline{\delta}} \overline{u}d\overline{y} . \quad (16)$$

Therefore, the feature of the resulting model is the van Driest [8] distribution modified in the outer region by the Lamourelle and

Sandall [5] model. It is a common practice to use a constant turbulent Schmidt number in the boundary layer analysis and for the present analysis $Sc_t = 1$ was used.

The overall mass transfer coefficient from the interface to the inlet liquid film is defined as:

$$k_c = \frac{\int_0^z D \frac{\partial C}{\partial y}|_\delta dz}{(C_s - 0)} .$$ (17)

where C_s is the surface concentration. Sherwood number can be calculated from the following equation:

$$Sh = \frac{kz}{D} \frac{\int_0^{\bar{z}} \frac{\partial \overline{C}}{\partial \overline{y}}|_{\overline{y} = \overline{\delta}} d\overline{z}}{\overline{C}_s} .$$ (18)

The overall dimensionless mass transfer rate, $\overline{m}$ is:

$$\overline{m} = \int_0^{\overline{z}} \frac{\partial \overline{C}}{\partial \overline{y}}|_{\overline{y} = \overline{\delta}} d\overline{z} .$$ (19)

The differential Equation 7 was put in a finite difference form using implicit scheme. The Crank-Nicolson method was used for Equation 7, which yielded a three-diagonal matrix. Four hundred increments were chosen in the $\overline{y}$ direction and, using a step-by-step advancing technique, $\overline{C}$ was calculated at different values of $\overline{z}$.

RESULTS AND DISCUSSION

The numerical values which were used in our calculations were: $\nu = 8.63 \times 10^{-7} \frac{m^2}{sec}$, $\rho = 996.3 \frac{kg}{cm^3}$, $D = 1.95 \times 10^{-9} \frac{m^2}{sec}$, $Sc = 442$, $g = 9.81 \frac{m}{sec^2}$, and $\sigma = 0.0689 \frac{N}{m}$. In most of our calculations, we used $\overline{\delta} = 100$, which is equivalent to the Reynolds number of 5000. For the physical absorption ($\alpha = 0$), development of the concentration profile is shown in Figure 2. Calculations were carried up to $\overline{z} = 10^6$, which is equivalent to the column length of $z = 7.24$ m. For the case of laminar flow, development of the concentration profile for physical absorption for $\overline{\delta} = 6.66$ (Re = 59.1) is shown in Figure 3. The relation between two
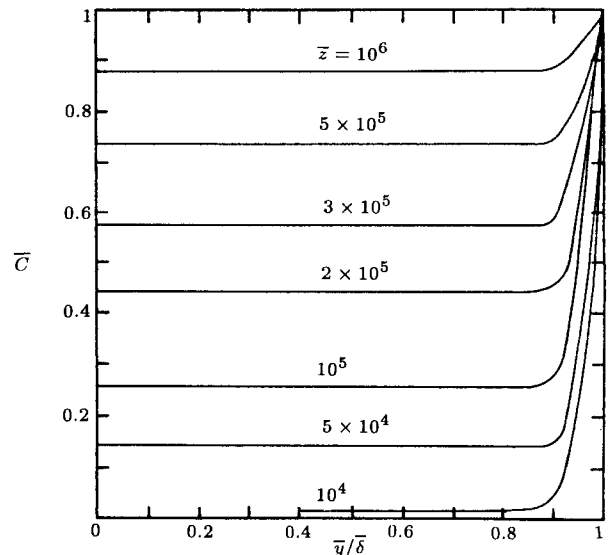

Figure 2. Development of the concentration profile for no resistance in the gas phase and without chemical reaction for $\overline{\delta} = 100$ (Re = 5000).
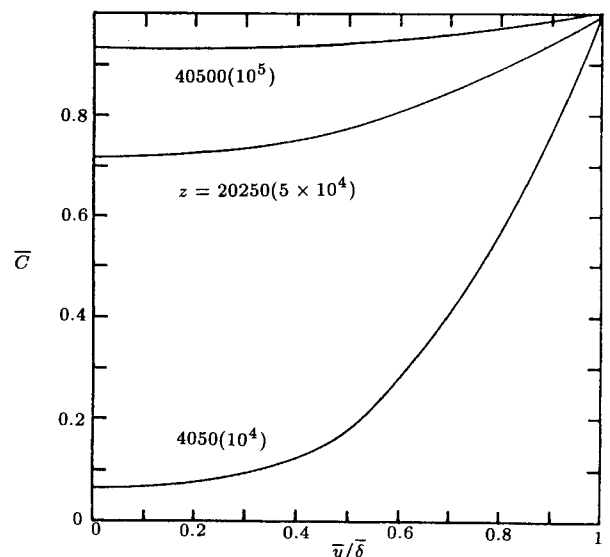

Figure 3. Development of the concentration profile for no resistance in the gas phase and without chemical reaction for $\overline{\delta} = 6.66$ (Re = 59.1).

different values of $\overline{\delta}_1$ and $\overline{\delta}_2$ is:

$$\frac{\overline{z}_1}{\overline{z}_2} = (\frac{\overline{\delta}_1}{\overline{\delta}_2})^{\frac{1}{3}} .$$

Thus:

$$\overline{z}(\overline{\delta} = 6.66) = 0.405\overline{z}(\overline{\delta} = 100) .$$

In Figure 3, values of $\overline{z}$ correspond to the same dimensional axial distance z as in
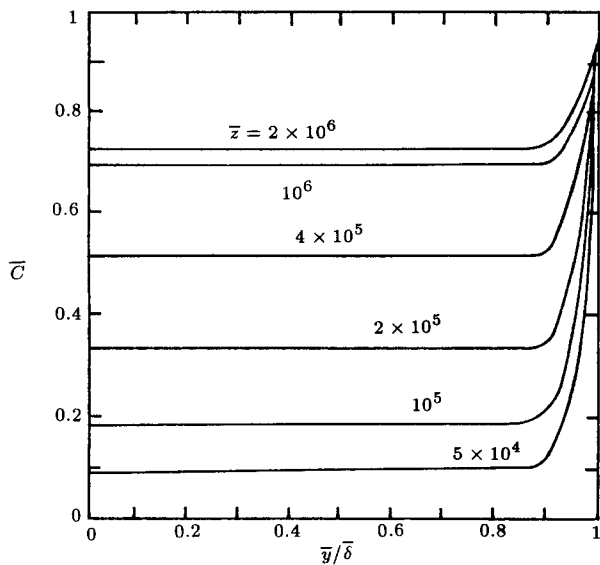
Figure 4. Development of the concentration profile for no resistance in the gas phase and without chemical reaction for $\bar{\delta} = 200$ (Re $= 11383$).
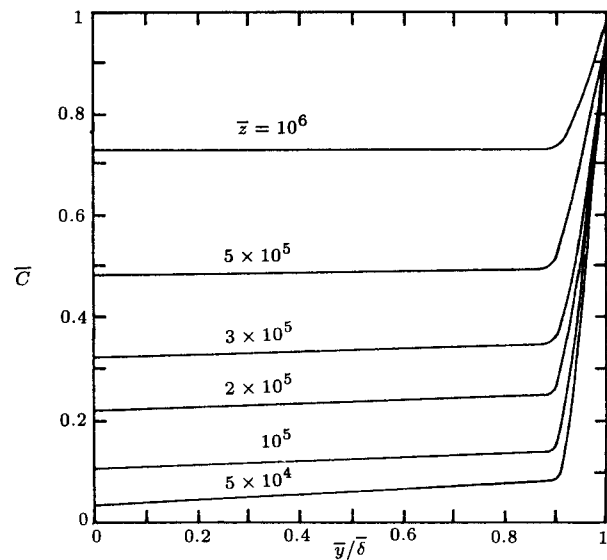

Figure 5. Development of the concentration profile for no resistance in the gas phase and $\alpha = 1$ for $\bar{\delta} = 100$.

Figure 2. Results shown in Figure 3 are in good agreement (less than 0.5% deviation) with the analytical solution of Olbricht and Wild [1]. Figure 4 shows the same result for turbulent flow when $\bar{\delta} = 200$(Re $= 11383$). Comparison of Figures 2 and 3 indicates that turbulent flow concentration is less than in laminar flow, while the slope of $\frac{\partial \bar{C}}{\partial \bar{y}}$ at free surface in turbulent is greater than in laminar flow. Therefore, as expected, the rate of gas absorption in turbulent flow is much higher than that in laminar flow.
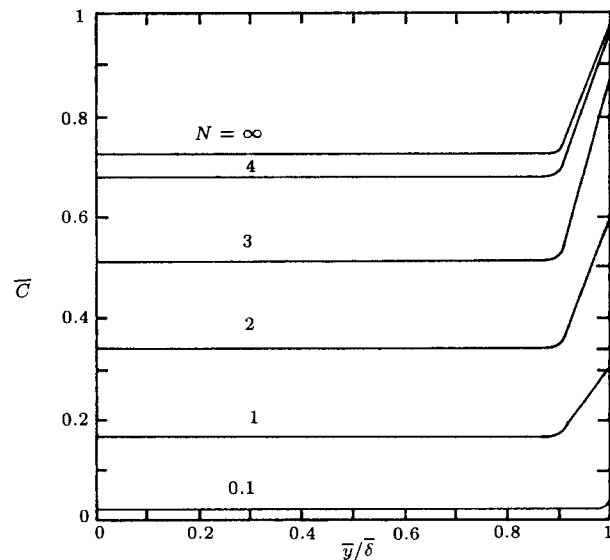
The effect of chemical reaction on the concentration profile is shown in Figure 5. Comparison of Figures 5 and 2 shows how presence of a reaction reduces concentration of absorbing spieces. The effect of mass transfer resistance (N) on the concentration profile at $\bar{z} = 10^6$ is shown in Figure 6. As N decreases, concentration decreases too. A graphical relation between $\bar{\delta}$ and Re is shown in Figure 7. Transition from laminar to turbulent flow is at Re $= 1200 - 1300$, for laminar flow, Re $= (\frac{4}{3})\bar{\delta}^2$ and for turbulent flow for values of $\bar{\delta} > 60$, Re can be related to $\bar{\delta}$ by a linear relation of Re $= 63.83\bar{\delta} - 1383$ with less than 1% deviation. Mass transfer rates (m) are

Figure 6. Concentration profile with gas phase resistance and $\alpha = 1$ for $\bar{\delta} = 100$ at $\bar{z} = 10^6$.

shown in Figure 8 for special cases. The two lower curves represent rates of absorption for laminar films $(\bar{\delta} = 6.66$ or Re $= 59.1)$ for $\alpha = 0$ (physical absorption alone) and $\alpha = 1$ respectively. For turbulent flow $(\bar{\delta} = 100$, Re $= 5000)$ the overall mass transfer rates are shown for $\alpha = 0, 1$, and 10. As can be seen, the effect of the chemical reaction on the absorption rate in the laminar film is greater than that in the turbulent flow. Absorption rate for
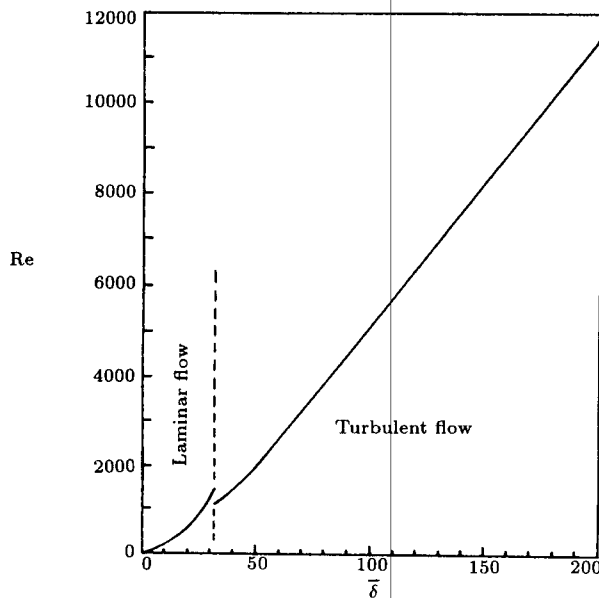
Figure 7. Relation between the dimensionless film thickness, $\bar{\delta}$, and Reynolds number.
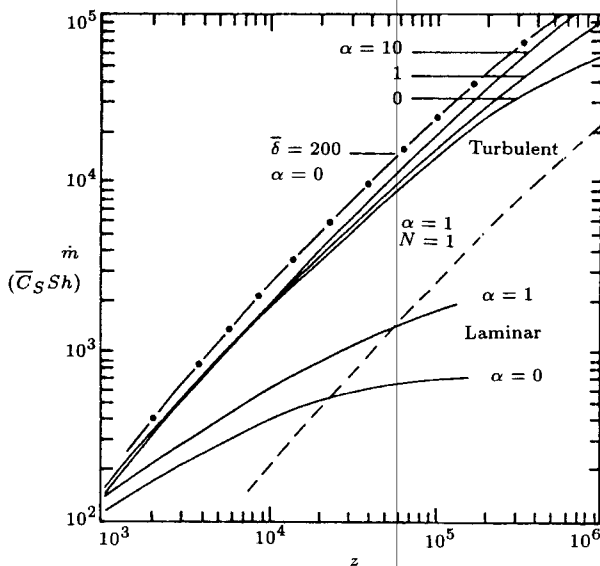

Figure 8. Gas absorption rates in laminar and turbulent films with and without chemical reaction.

$N = 1$ and $\alpha = 1$ is also shown in Figure 8. For higher Reynolds numbers ($\delta = 200$ or Re $= 11383$), the absorption rate for $\alpha = 0$ is also shown in Figure 8. It should be noted that in Figure 8, for different dimensionless film thickness, $\bar{\delta}$, corresponding values of $\bar{z}$ for same dimensionless axial distance $\bar{z}$ are different. In fact for $\bar{\delta} = 6.66$; $\bar{z}(\bar{\delta} = 6.66) = 0.405\bar{z}(\bar{\delta} = 100)$ and for $\bar{\delta} = 200$, $\bar{z}(\bar{\delta} = 200) = 1.26\bar{z}(\bar{\delta} = 100)$.

CONCLUSIONS

The numerical solution of gas absorption into laminar and turbulent falling films, with the combined effects of homogeneous, irreversible first-order chemical reaction and gas phase mass-transfer resistance, has been presented. Results are given in the forms of concentration profile development and the rate of gas absorption. It has been shown that the relative effect of the chemical reaction on the absorption rate in the laminar flow is greater than that in the turbulent flow, while the absolute mass-transfer rates in turbulent-case are significantly larger than those observed in laminar flow. It has been shown that in turbulent flow, when Reynolds number or the film thickness increases, absorption rates also increase. With the gas phase mass-transfer-resistance, the absorption rates can be significantly smaller than those for the case of no resistance, particularly when the parameter N is small.

NOMENCLATURE

A	damping length constant $(26\nu\frac{\rho}{\tau_0})$
C	concentration of dissolved gas in the liquid
C^*	equilibrium concentration of gas in liquid
C_s	concentration of gas at the free surface
D	molecular diffusion coefficient
g	acceleration of gravity
g_c	gravity constant (1 in SI unit system)
k	first-order reaction rate constant
k_c	mass transfer coefficient in liquid phase
k_G	mass transfer coefficient in the gas phase
K'	constant (0.4)
K''	constant
L'	Prandtl mixing length
$\bar{m}$	dimensionless rate of absorption

N dimensionless parameter ($\frac{k_G}{D}[\frac{\nu^2}{g}]^{\frac{1}{3}}$)

Re   Reynolds number

Sc Schmidt number ($\frac{\nu}{D}$)

Sc_t turbulent Schmidt number ($\frac{\varepsilon_M}{\varepsilon_D}$)

Sh Sherwood number ($\frac{k_c z}{D}$)

u liquid velocity in axial direction

y distance normal to the surface

z axial distance

Greek Letters

α reaction rate constant ($k(\frac{\nu}{g^2})^{\frac{1}{3}}$)

Γ liquid loading

δ liquid film thickness

ε_D eddy diffusivity

ε_M kinematic eddy viscosity

σ surface tension

ρ liquid density

μ liquid viscosity

ν liquid kinematic viscosity

τ_o shear stress at the free surface

Superscript

$-$ nondimensionalized quantity

AKNOWLEDGEMENTS

REFERENCES

1. Olbrich, W.E. and Wild, J.D. "Diffusion from the free surface into a liquid film in laminar flow over defined shapes", *Chem. Engng. Sci.*, **24**(25) (1969).

2. Best, R.J. and Hoerner. "Gas absorption with first-order chemical reaction in laminar falling films – calculation of rates and enhancement factors", *Chem. Engng. Sci.*, **34**, pp 759-762 (1979).

3. Riazi, M.R. "Estimation of rates and enhancement factors in gas absorption with chemical reaction and gas phase mass transfer resistances", *Chem. Eng. Sci.*, **41**(11), p 2925 (1986).

4. Riazi, M.R. and Fari, A. "Effect of interfacial drag on gas absorption with chemical reaction", *AIChE J.*, **32**(4), p 696 (1986).

5. Lamourelle, A.P. and Sandall, O.C. "Gas absorption into a turbulent liquid", *Chem. Engng. Sci.*, **27**, pp 1035-1043 (1972).

6. Menez, G.D. and Sandall, O.C. "Gas absorption accompanied by first-order chemical reaction in turbulent liquids", *Ind. Eng. Chem. Fundam.*, **13**(1), pp 72-76 (1974).

7. Seban, R.A. and Fari, A. "Heating and evaporation with turbulent falling liquid film", *J. of Heat Transfer*, **98** (1976).

8. Van Driest, E.R. "On turbulent flow near the wall", *J. Aero. Sci.*, **23**, p 1007 (1956).

9. Levich, V.G. *Physiochemical Hydrodynamics*, Prentice Hall, Englewood Cliffs, New Jersey, p 691 (1962).