

Gas-Liquid Mass Transfer in Taylor Flow Through a Capillary

SAID IRANDOUST, SYLVIE ERTLÉ* and BENGT ANDERSSON[#]

Department of Chemical Reaction Engineering, Chalmers University of Technology, S-412 96 Gothenburg, Sweden

The gas-liquid mass transfer in two-phase flow through a capillary has been measured for water-air, ethanol-air and ethylene glycol-air systems. A semi-theoretical model has been developed and compared with experimental results, and a full computer simulations of the flow pattern and mass transfer using a flow simulation program have been made. The measured values are about 30% less than the calculated values.

On a mesuré le transfert de matière gaz-liquide dans un écoulement biphasique dans un capillaire pour des systèmes eau-air, éthanol-air et éthylène glycol-air. Un modèle semi-théorique a été mis au point et comparé avec les résultats expérimentaux, et des simulations du profil d'écoulement et du transfert de matière ont été effectuées par ordinateur à l'aide d'un programme de simulation d'écoulement. Les valeurs mesurées sont environ 30% inférieures aux valeurs calculées.

Keywords: Taylor flow, segmented flow, mass transfer, monolith.

In many three-phase systems, where gas and liquid compounds react in the presence of a solid catalyst, the mass transfer rate between phases is important for efficient design of the reactor. During the preceding decade, monolithic catalyst reactors have been used successfully in chemical technology (Irandooust and Andersson, 1988a). The monoliths are composed of a large number of axial porous channels through which gas and liquid flow co-currently. For two-phase flow, the segmented gas-liquid flow (Taylor flow) in the monolithic catalyst reactor gives superior mass transfer characteristics of the reactor (Irandooust and Andersson, 1989a).

In Taylor flow the liquid slugs are separated by large bubbles the length of which is greater than the channel diameter. The gas bubbles (Taylor bubbles) are separated from the wall by a thin liquid film causing a very low axial dispersion between liquid slugs. The bubbles also prevent the development of parabolic flow in the liquid slugs, and this leads to recirculation. The recirculation within the liquid slugs enhances the radial mass transfer considerably. Figure 1 shows the flow pattern which occurs in Taylor flow in the monolith channels.

Other advantages resulting from the application of Taylor flow in the monolithic structure are low pressure drop, high surface area, uniformity of the distribution of the flow, short diffusion length in the solid catalyst and nearly direct contact between the wall and the gas.

The features of this flow type and its application have been studied by Irandooust and Andersson (1988b, 1989a) and Irandooust et al. (1989). The purpose of this work is to describe the gas-liquid mass transfer phenomena occurring in a single circular channel of the monolith. The flow type studied is upward Taylor flow. Most investigations devoted to mass transfer in two-phase flow are carried out with tubes of larger diameters than those of interest for a monolithic structure. Recently, Kawakami et al. (1989) have investigated gas-liquid and liquid-solid mass transfer rates in a monolithic reactor with different channel sizes. The authors have demonstrated the superior mass transfer properties of the monolithic reactor compared with a packed-bed reactor. Van Heuven and Beek (1963) studied the absorption of carbon dioxide into water under slug conditions in gas lifts. The liquid phase resistance to mass transfer was calculated for a single bubble.

Votruba et al. (1975) studied mass transfer from gas to solid by the vapourisation of water or hydrocarbons from the porous surface of monoliths. They correlated their data to Reynolds and Schmidt numbers. Other studies in this area though for tubes of larger diameters, have been carried out by Tomida et al. (1978), Shilimkan and Stepanek (1977a,b), Kasturi and Stepanek (1974a,b) and Infante Ferreira (1985). Furthermore, the solid-liquid mass transfer in Taylor flow has been investigated by Vrentas et al. (1978); Hatziantoniou and Andersson (1982) and Edvinsson and Irandooust (1992).

Experimental

A schematic diagram of the experimental equipment is shown in Figure 2. Gas and liquid were introduced to the glass capillary through the inlet cell where gas and liquid slugs were formed. The glass capillaries used were of 1.5 and 2.2 mm inner diameter and 0.60 m long. The slug lengths of gas and liquid could be varied independently and were measured by use of a photocell enclosing the glass tube. The lengths of the gas slugs varied from 3.4 to 29.1 mm and the lengths of the liquid slugs varied from 2.9 to 67 mm. The values of average total linear velocity were determined to be from 0.092 to 0.56 m/s, corresponding to Reynolds numbers ranging from 11 to 824, respectively.

The gas-liquid mass transfer experiments were carried out by absorption of oxygen from air into three different liquids possessing different physical properties. The liquids chosen were water, ethanol and ethylene glycol. The physical properties of these liquids are listed in Table 1. The experimental procedure was as follows: The degassed liquid (saturated with nitrogen) and air were fed to the inlet cell at desired flow rates. After passage through the glass capillary, the gas and liquid slugs were separated at the outlet cell. An oxygen electrode (OXI 530, EO 200, by Wissenschaftlich Technische Werkstätten, Weilheim Germany) placed in the liquid passage measured the oxygen concentration in the outgoing liquid. This method of measurement revealed the total rate of gas-liquid mass transfer occurring in the inlet cell, the glass capillary, and in the outlet cell. The contributions of the inlet and outlet cells to the total mass transfer were eliminated by undertaking two sets of measurements with capillaries of different lengths. The mass transfer rate occurring in the capillary itself was calculated from the difference between the value obtained for the whole set-up consisting of the inlet cell, the capillary and the outlet cell, and the value of mass transfer in an arrangement in

*Present address: Millipore, Zone industrielle, F-20 671, Molsheim, France.

[#]Correspondence: Dr. Bengt Andersson.

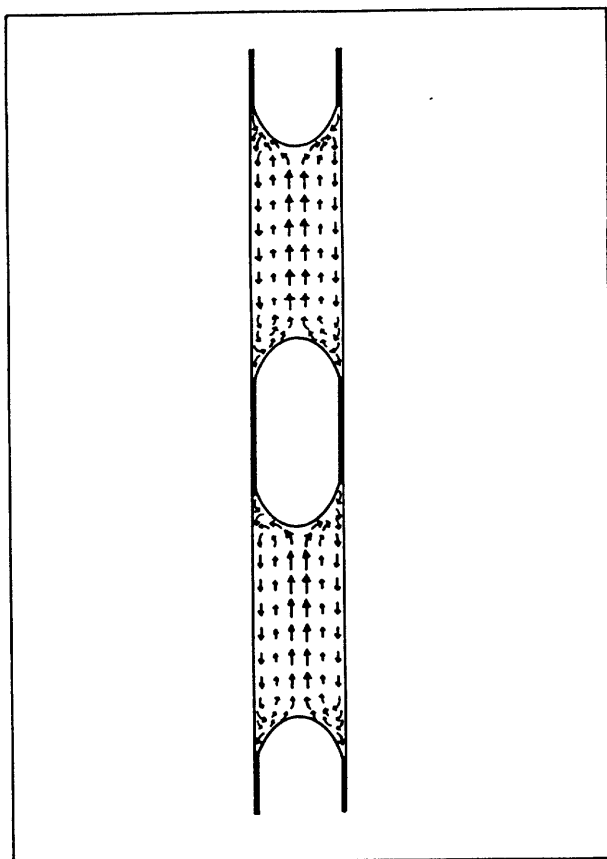


Figure 1 — Flow pattern in vertical Taylor flow.

which the inlet and outlet cells were connected by a short piece of the capillary. All runs were performed at atmospheric pressure and room temperature (20–25°C). The resistance to mass transfer in the gas phase was negligible due to the relatively high gas phase diffusivity of oxygen and slow interphase mass transfer rate.

Theory

A semi-theoretical model for correlating the mass transfer coefficients was developed. The interpretations of this correlation have been checked by use of computer simulations of gas-liquid mass transfer.

SEMI-THEORETICAL MODEL

It was assumed that the gas slugs were cylindrical with hemi-spherical top and bottom menisci surfaces, and that they were well separated by liquid slugs. The mass transfer occurring from a single Taylor bubble was assumed to occur partly from the cylindrical part into the liquid film and partly through the top and bottom menisci surfaces. These contributions were calculated theoretically and empirically.

At the cylindrical part of the Taylor bubble, the liquid film falls down between the gas slug and the capillary wall. The steady state parabolic velocity profile for the film is given by Skelland (1974) as

$$u(x) = (\rho g \delta^2 / 2\mu) [1 - (x/\delta)^2] \quad (1)$$

The film thickness δ in Equation (1) was calculated from the empirical relationship given by Irandoust and Andersson (1989b):

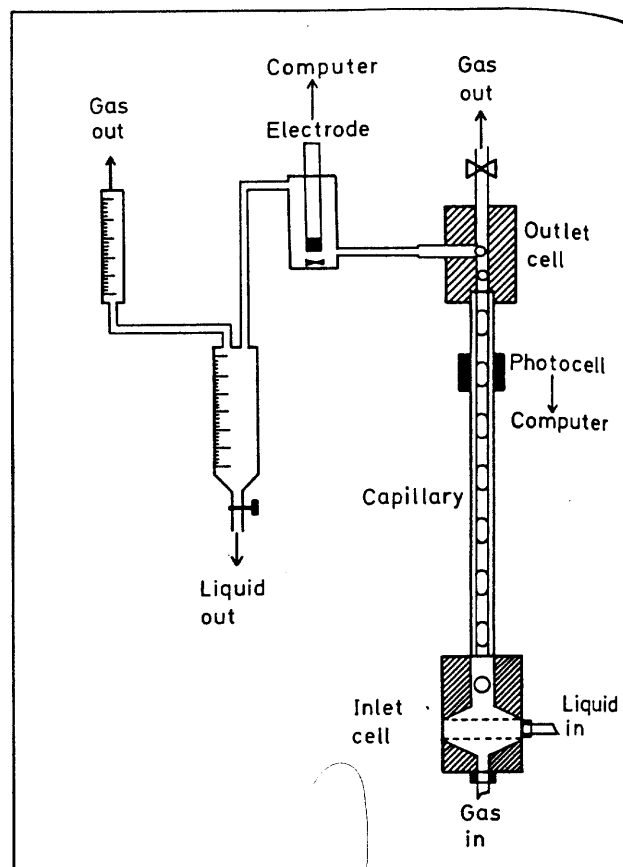


Figure 2 — Experimental set-up.

TABLE 1
Properties of the Liquids Used at 20°C

	$\mu \cdot 10^3$ (kg/m · s)	ρ (kg/m ³)	$\sigma \cdot 10^3$ (N/m)	$D \cdot 10^9$ (m ² /s)
Water	1.0	998	72.8	2.10
Ethanol	1.4	809	29.9	2.13
Ethylene Glycol	21.1	1121	46.3	0.26

$$\delta = 0.18 d_i \{1 - \exp[-3.1 (\mu v / \sigma)^{0.54}]\} \quad (2)$$

It should be noted that the liquid film inherent in Taylor flow is very thin for low and medium viscosity liquids. Also the ratio of the film penetration distance to the capillary radius is small due to the very short time of exposure between the Taylor bubble and the renewed liquid film. In such a case rectangular coordinates can be applied in place of cylindrical coordinates (see Appendix A).

A mass balance for the dissolved oxygen over a differential element of length dz at distance x from the gas-liquid interface (Figure 3) may be written as (Bird et al., 1960)

$$u(x)(\partial c / \partial z) = D(\partial^2 c / \partial x^2) \quad (3)$$

using rectangular coordinates. Here it is assumed that the solute moves in the z direction primarily by convective motion with the diffusive contribution being negligible, while in the x direction mass transfer is due mainly to diffusion. The resistance to mass transfer in the gas phase was not considered. The partial differential equation is to be solved with the following boundary condition:

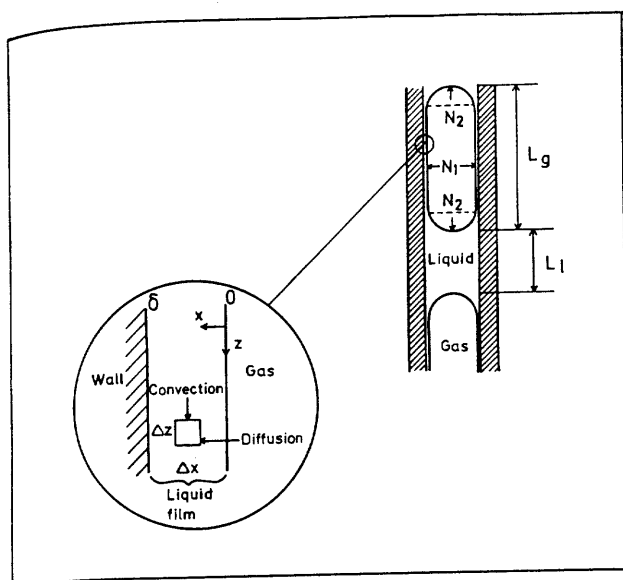


Figure 3 — Diffusion into a falling film.

$$c = c_0 \quad \text{at } z = 0, \text{ all } x \quad (4)$$

$$c = c^* \quad \text{at } z > 0, x = 0 \quad (5)$$

$$(\partial c / \partial x) = 0 \quad \text{at all } z, x = \delta \quad (6)$$

where c^* is the concentration of dissolved gas in equilibrium with the gas itself, and c_0 is the initial concentration of the solute in the liquid. In order to make the solution of Equation (3) easier, a coordinate system which moves with the slugs was introduced. Hence, the total film velocity observed from the fixed Taylor bubble is

$$U(x) = u(x) + v \quad (7)$$

where v is the wall velocity when the slugs are fixed. Equation (3) can now be rewritten as

$$U(x)(\partial c / \partial z) = D(\partial^2 c / \partial x^2) \quad (8)$$

when the wall velocity, v , in Equation (7) is the dominating one, a solution of Equation (8) is (Jenson and Jeffreys, 1977):

$$\begin{aligned} [c(z, x) - c_0] / [c^* - c_0] = \sum_{n=0}^{\infty} (-1)^n \\ \{ \operatorname{erfc} [(x + 2n\delta) / (4Dz/v)^{0.5}] \\ + \operatorname{erfc} [(2n\delta + 2\delta - x) / (4Dz/v)^{0.5}] \} \quad (9) \end{aligned}$$

The mixed cup concentration of the solute in the liquid film, being in contact with the cylindrical part of the Taylor bubble, is given by:

$$\begin{aligned} \frac{c_m - c_0}{c^* - c_0} = \left\{ \int_0^\delta \left[\frac{2(R - \delta + x)}{2R - \delta} \right] \right. \\ \left. \left[\frac{c(z = L_c, x) - c_0}{c^* - c_0} \right] dx \right\} / \delta = y_m \quad (10) \end{aligned}$$

The average mass transfer into the liquid film, from the cylindrical part of the Taylor bubble can be written as

$$N_1 = (c_m - c_0) U_{av} A_1 \quad (11)$$

where U_{av} is the average velocity of the liquid film over a cross section of the film and is obtained with the aid of Equations (1) and (7) as

$$U_{av} = (\rho g \delta^2 / 3\mu) + v \quad (12)$$

The cross section area of the film is

$$A_1 = \pi \delta (d_t - \delta) \quad (13)$$

Hence,

$$N_1 = [\pi \delta (d_t - \delta) U_{av} y_m] (c^* - c_0) \quad (14)$$

The mass transfer from the spherical ends of the Taylor bubbles is calculated by empirical relations, using the Sherwood number (Clift et al., 1978)

$$\begin{aligned} (Sh - 1) / Sc^{1/3} = [1 + (1/ReSc)^{1/3} Re^{0.41}] \\ 1 \leq Re \leq 400 \quad (15) \end{aligned}$$

and

$$\begin{aligned} Sh = 1 + 0.724 Re^{0.48} Sc^{1/3} \quad 100 \leq Re \leq 2000 \\ \quad (16) \end{aligned}$$

Equations (15) and (16) were derived for rigid spheres. Irandoost and Andersson (1989b) recently have shown that the spherical ends of the Taylor bubbles can be treated as rigid spheres. Hence, the liquid side mass transfer coefficient for this part, $k_{L,2}$, can be written as

$$k_{L,2} = (D Sh / d_B) \quad (17)$$

Now, the contribution of spherical ends to the total mass transfer is given by

$$N_2 = k_{L,2} A_2 (c^* - c_0) \quad (18)$$

The transfer surface area is

$$A_2 = \pi d_B^2 \quad (19)$$

resulting in

$$N_2 = (\pi D Sh d_B) (c^* - c_0) \quad (20)$$

The total gas-liquid mass transfer can now be obtained by adding Equations (14) and (20);

$$\begin{aligned} N_{tot} = N_1 + N_2 = \{ \pi [\delta (d_t - \delta) U_{av} y_m \\ + D Sh d_B] \} (c^* - c_0) \quad (21) \end{aligned}$$

where the quantity in large brackets divided by the volume of the element under consideration is the semi-theoretical mass transfer coefficient:

$$k_L a = 4 [\delta (d_t - \delta) U_{av} y_m + D Sh d_B] / [d^2 (L_1 + L_g)] \quad (22)$$

COMPUTER SIMULATIONS

The gas-liquid and gas-liquid-solid mass transfer with very fast chemical reaction on the solid wall, have been simulated using a finite difference program (PHOENICS by CHAM Ltd).

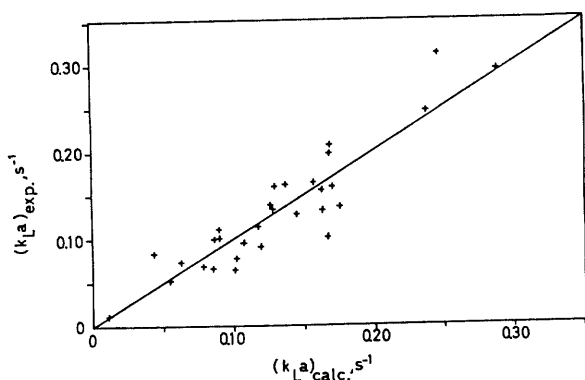


Figure 4 — Experimental results: volumetric mass transfer coefficient.

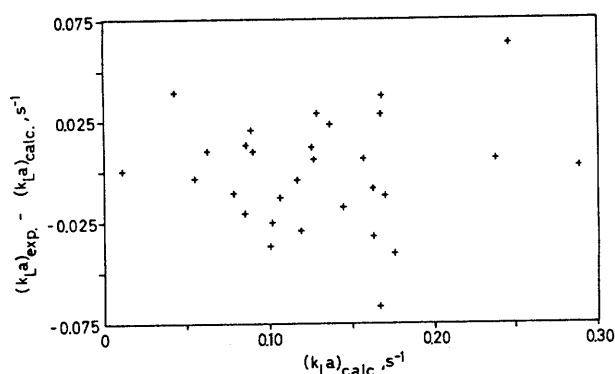


Figure 5 — Residuals plotted against fitted values from Equation (23).

The flow equations and boundary conditions were solved for a fraction of the tube containing two Taylor bubbles and two liquid slugs. More details about this part can be found elsewhere (Irandoost and Andersson, 1989a).

Results and discussion

In order to measure the gas-liquid mass transfer in Taylor flow, thirty runs were performed. These experiments were carried out using different liquids, different flow rates and two different tube sizes. In the present work the semi-theoretical formula

$$(k_L a)_{calc.} = \alpha (k_L a) \quad (23)$$

was fitted to the experimental $(k_L a)$ values by regression analysis. The value of the fitted parameter, with a 95% confidence interval, is 0.686 ± 0.048 . Figure 4 shows the experimental $(k_L a)$ values versus calculated ones using Equation (23). In Figure 5, the residuals are plotted against the predicted $(k_L a)$ calculated from Equation (23). The 'horizontal band' of values predicted indicates no abnormality. In spite of the deviation of the parameter α from unity, the facts that the residuals are randomly distributed and that the model fits for all three liquids show that the theoretical model developed, Equation (22) is correct in form. The parameter deviation can be due to impurities that may arise in the liquid, making the whole liquid surface more rigid. The presence of such impurities substantially reduces the rate of mass transfer due to their accumulation at the gas-liquid interface (Skelland, 1974). These effects are highly dependent on both the structures and concentrations of the impurities, making any attempt to take this into consideration impractical.

The computer simulations of mass transfer were performed for a water-air system with and without reaction at the wall. It was shown that the mixing of the liquid film into the succeeding liquid slug was incomplete. Since complete mixing was assumed in the theoretical model, the mass transfer rate will be overestimated. It was also concluded that both the cylindrical part and the rear ends of the gas slugs made almost the same contribution to the gas transfer in terms of mole/s m^2 transfer area. This finding contradicts the results of Van Heuven and Beek (1963) who studied carbon dioxide absorption into water under slug conditions. Computer simulations of a fast reaction occurring on a solid wall show that even during these conditions the mass transfer rate on the cylindrical and the spherical parts of the bubble are about the same, expressed as mole/s m^2 . However, since the surface area of the cylindrical part is usually 5–10 times larger, this part is most important. For a fast reaction occurring on a solid wall, the concentration profile in the liquid film reaches a stationary value for a thin film and long bubbles. In such a case the mass transfer coefficient can with good accuracy be approximated by

$$k_L = D/\delta \quad (24)$$

The influence of the liquid film on the mass transfer coefficient can be clarified by an analysis of the concentration profile of the solute in the liquid film. During the very short time when a liquid element in the liquid film is exposed to the air bubble, the solute does not penetrate through the whole liquid film to the wall, except in cases involving long gas slugs, thin film or low velocity.

Conclusions

The measured gas-liquid mass transfer coefficients for all the three liquids tested, i.e. water, ethanol and ethylene glycol, agree very well with calculated values based on the developed theoretical approximation.

Gas-liquid mass transfer in Taylor flow in a capillary is mainly determined by the liquid-phase diffusion coefficient and the gas-liquid contact area. The flow rate and the gas and liquid slug lengths have a minor influence. The gas-liquid contact area is affected only by the channel diameter and gas hold-up and is easy to calculate. This makes it possible to calculate the mass transfer coefficient, within a factor of two, without measurements in the actual system.

The total mass transfer is high due to the high gas-liquid and liquid-solid contact area (3000–5000 m^2/m^3 reactor).

Acknowledgements

The authors wish to acknowledge the financial support of the Swedish National Board of Technical Development and the National Energy Administration of Sweden.

Nomenclature

- a = radius, gas slug, m
- A = parameter defined by Equation (A3)
- A_1 = cross section area, liquid film, m^2
- A_2 = transfer area, spherical ends, m^2
- B = parameter defined by Equation (A4)
- c = solute concentration, mol/m^3
- c_0 = solute concentration, initial, mol/m^3
- c_m = solute concentration, mixed-cup, mol/m^3
- c^* = saturation concentration, mol/m^3
- C = parameter defined by Equation (A5)

d_B = diameter, Taylor bubble, m
 d_t = diameter, tube, m
 D = liquid-phase diffusion coefficient, m^2/s
 g = acceleration due to gravity, m/s^2
 k_L = mass transfer coefficient, spherical ends, m/s
 $k_{L,2}$ = mass transfer coefficient, s^{-1}
 $k_{L,a}$ = volumetric mass transfer coefficient, s^{-1}
 L_c = length of the cylindrical part of Taylor bubble, m
 L_g = slug length, gas, m
 L_l = slug length, liquid, m
 N_1 = mass transfer through the film, mol/s
 N_2 = mass transfer from the spherical ends, mol/s
 r = radial distance, m
 R = tube radius, m
 Re = Reynolds number = $d_t v \rho / \mu$
 Sc = Schmidt number = $\mu / \rho D$
 Sh = Sherwood number = $k_L d_B / D$
 t = exposure time of gas slug, s
 u = velocity of free-falling liquid film, m/s
 U = total velocity in liquid film, m/s
 U_{av} = average total velocity in liquid film, m/s
 v = wall velocity, m/s
 x = distance from the gas-liquid interface, m
 x_p = film penetration distance, m
 y_m = constant defined by Equation (10)
 z = axial distance, m

Greek symbols

α = regression parameter in Equation (23)
 β = constant defined by Equation (A2)
 δ = liquid film thickness, m
 μ = liquid viscosity, $kg/m \cdot s$
 ρ = liquid density, kg/m^3
 σ = air-liquid surface tension, N/m

APPENDIX A

The mass transfer equation using cylindrical coordinates is:

$$D[(\partial^2 c / \partial r^2) + (1/r)(\partial c / \partial r)] = v(\partial c / \partial z) \quad (A1)$$

According to Crank (1975), if the initial concentration is c_0 and the concentration at the bubble surface is c^* (at $r = a$, where $a = R - \delta$) then

$$\beta = (c - c_0) / (c^* - c_0) = (a/r)^{0.5} \operatorname{erfc}(A) + B \operatorname{ierfc}(A) + C i^2 \operatorname{erfc}(A) + \dots \quad (A2)$$

where

$$A = (r - a) / (4 D t)^{0.5} \quad (A3)$$

$$B = (r - a)(D t)^{0.5} / (4 a^{0.5} r^{1.5}) \quad (A4)$$

$$C = (D t)(9a^2 - 2ar - 7r^2) / (32 a^{1.5} r^{2.5}) \quad (A5)$$

for small times.

The mixed cup concentration is given by

$$y_m = (1/\delta) \int_a^R [2r / (2R - \delta)] \beta dr \quad (A6)$$

We can also calculate penetration distance as

$$x_p = (D t)^{0.5} \quad (A7)$$

where t is the time of exposure of gas slug and can be obtained as

$$t = L_g / v \quad (A8)$$

The time of exposure of gas slugs is under 0.1 s in all our experiments. Hence the ratio (x_p / R) is always below 0.02 in the present work.

A comparison of y_m values obtained by applying Equation (A6) and Equation (10) showed a maximum difference of about 2%. Hence the error obtained in assuming rectangular coordinates instead of cylindrical coordinates is within the experimental accuracy and should be acceptable.

References

- Bird, R. B., W. E. Stewart and E. N. Lightfoot, "Transport Phenomena", John Wiley & Sons, New York (1960).
- Clift, R., J. R. Grace and M. E. Weber, "Bubbles, Drops, and Particles", Academic Press, New York (1978).
- Crank, J., "The Mathematics of Diffusion", Clarendon Press, Oxford (1975).
- Edvinsson, R. and S. Irandoust, To be published (1992).
- Hatziantoniou, V. and B. Andersson, "Solid-Liquid Mass Transfer in Segmented Gas-Liquid Flow through a Capillary", Ind. Eng. Chem. Fundam. **21**, 451-456 (1982).
- Infante Ferreira, C. A., "Combined Momentum, Heat and Mass Transfer in Vertical Slug Flow Absorbers", Int. J. Refrig. **8**, 326-334 (1985).
- Irlandoust, S. and B. Andersson, "Simulation of Flow and Mass Transfer in Taylor Flow through a Capillary", Comput. Chem. Eng. **13**, 519-526 (1989a).
- Irlandoust, S. and B. Andersson, "Liquid Film in Taylor Flow through a Capillary", Ind. Eng. Chem. Res. **28**, 1684-1688 (1989b).
- Irlandoust, S. and B. Andersson, "Monolithic Catalysts for Non-automobile Applications", Catal. Rev. Sci. Eng. **30**, 341-392 (1988a).
- Irlandoust, S. and B. Andersson, "Mass Transfer and Liquid-Phase Reactions in a Segmented Two-Phase Flow Monolithic Catalyst Reactor", Chem. Eng. Sci. **43**, 1983-1988 (1988b).
- Irlandoust, S., B. Andersson, E. Bengtsson and M. Siverström, "Scaling Up of a Monolithic Catalyst Reactor with Two-Phase Flow", Ind. Eng. Chem. Res. **28**, 1489-1493 (1989).
- Jenson, V. G. and G. V. Jeffreys, "Mathematical Methods in Chemical Engineering", Academic Press, London (1977).
- Kasturi, G. and J. B. Stepanek, "Two-Phase Flow-III. Interfacial Area in Co-current Gas-Liquid Flow", Chem. Eng. Sci. **29**, 713-719 (1974a).
- Kasturi, G. and J. B. Stepanek, "Two-Phase Flow-IV. Gas and Liquid Side Mass Transfer Coefficients", Chem. Eng. Sci. **29**, 1849-1856 (1974b).
- Kawakami, K., K. Kawasaki, F. Shiraishi and K. Kusunoki, "Performance of a Honeycomb Monolith Bioreactor in a Gas-Liquid-Solid Three-Phase System", Ind. Eng. Chem. Res. **28**, 394-400 (1989).
- Shilimkan, R. V. and J. B. Stepanek, "Interfacial Area in Co-current Gas-Liquid Upward Flow in Tubes of Various Size", Chem. Eng. Sci. **32**, 149-154 (1977a).
- Shilimkan, R. V. and J. B. Stepanek, "Effects of Tube Size on Liquid Side Mass Transfer in Co-Current Gas-Liquid Upward Flow", Chem. Eng. Sci. **32**, 1397-1400 (1977b).
- Skelland, A. H. P., "Diffusional Mass Transfer", John Wiley & Sons, New York (1974).
- Tomida, T., F. Yusa and T. Okazaki, "Effective Interfacial Area and Liquid-Side Mass Transfer Coefficient in the Upward Two-Phase Flow of Gas-Liquid Mixtures", Chem. Eng. J. **16**, 81-88 (1978).
- Van Heuven, J. W. and W. J. Beek, "Gas Absorption in Narrow Gas Lifts", Chem. Eng. Sci. **18**, 377-390 (1963).
- Votruba, J., O. Mikus, K. Nguen, V. Hlavacek and J. Skrivaneck, "Heat and Mass Transfer in Honeycomb Catalyst-II", Chem. Eng. Sci. **30**, 201-206 (1975).
- Vrentas, S. J., J. L. Duda and G. D. Lehmkuhl, "Characteristics of Radial Transport in Solid-Liquid Slug Flow", Ind. Eng. Chem. Fundam. **17**, 39-45 (1978).

Manuscript received February 13, 1990; revised manuscript received September 21, 1990; accepted for publication January 7, 1991.