

## DIFFUSION MODEL OF GAS AND LIQUID MIXING IN BUBBLE COLUMNS

M. HARTMAN

*Institute of Chemical Process Fundamentals,  
Czechoslovak Academy of Sciences, 165 02 Prague - Suchbát*

Received January 15th, 1976

---

A diffusion model was employed to describe both the gas and the liquid mixing in bubble columns with countercurrent flow of the gas and liquid streams. Analytic solutions were developed that give concentration profiles within the bubble column for the case of the partial gas and liquid mixing in the vertical direction. From this a relation was developed and evaluated between the Murphree plate efficiency and the number of overall transfer units for different degrees of mixing in both phases.

---

A bubble column commonly used is a simple device in which the gas is dispersed into the liquid in a suitable manner. The net flow of the phases in the gas-liquid mixture can be counter- or co-current. The degree of liquid and gas mixing is one of the factors influencing the performance of such a gas-liquid contactor. When the gas and liquid flow rates are very low an assumption of piston flow of gas is usually made and seems to be feasible. Even in this quiescent regime the liquid is considerably mixed. At higher flow rates oscillations and circulations of the liquid may occur which generally tend to mix both the gas and liquid in columns. Such flow patterns often develop especially in large vessels. Knowledge the gas and liquid concentration gradients across the column and hence the degree of mixing in the both phases is important in the proper analysis of experimental data and the scaleup of contacting devices. While the effect of liquid mixing is taken into account in the design of bubble columns, piston flow of the gas phase is assumed<sup>1-3</sup>.

In this paper a diffusion model of liquid mixing<sup>4,5</sup> is extended to express quantitatively the relation between the column performance and the intensity of axial mixing in both the gas and the liquid.

### THEORETICAL

The diffusion model is based on the conception that the diffusion fluxes of a component in both phases are proportional to their concentration gradients. In analogy to molecular diffusion this constant is called turbulent diffusivity. This conception is justified in cases where the gas phase flows uniformly through the gas-liquid

mixture which is a reasonable first approximation for bubble columns under normal operating conditions. Although the mixing is rather caused by liquid circulation induced by the rising gas than a diffusive transport, we believe that the simplified model is a useful approach.

Mass transfer in a bubble column is treated here using the following simplifying assumptions: 1) uniform gas and liquid conditions in horizontal planes; 2) constant values of the overall mass transfer coefficient, turbulent diffusivities and the density of the gas-liquid mixture across the column; 3) counter-current flow of gas and liquid; 4) constant molar flow rates of each phase through the column; 5) linear gas-liquid equilibrium relation; 6) gas component is absorbed by liquid; 7) isothermal steady state operation.

A material balance over a differential element of bubble column as depicted in Fig. 1 leads to the relation

$$D_L F \rho_L (1 - e_G) \frac{d^2 x}{dH^2} - L \frac{dx}{dH} + D_G F \rho_G e_G \frac{d^2 y}{dH^2} + G \frac{dy}{dH} = 0. \quad (1)$$

If a chemical reaction occurs in either phase an additional reaction term appears in the balance. Only in the case of first (or zero) order reaction the analytical solution can be obtained. More complicated kinetics would require numerical integration<sup>8</sup>.

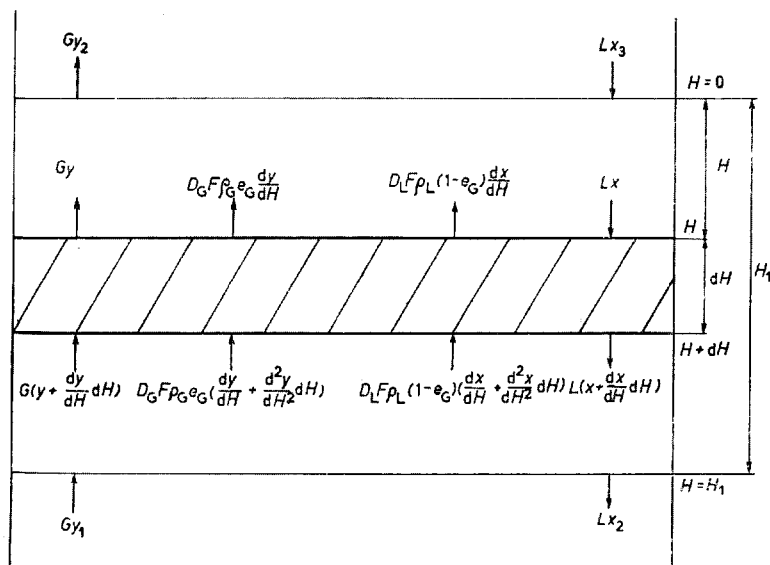


FIG. 1  
Diffusion Model of Gas and Liquid Mixing

Using the relative flow path  $w$  and the Peclet numbers for both phases this equation can be converted to dimensionless form

$$\frac{d^2x}{dw^2} - \text{Pe}_L \frac{dx}{dw} + \frac{G}{L} \frac{\text{Pe}_L}{\text{Pe}_G} \frac{d^2y}{dw^2} + \frac{G}{L} \text{Pe}_L \frac{dy}{dw} = 0, \quad (2)$$

where

$$\text{Pe}_G \equiv H_1 G / D_G F \varrho_G e_G \quad \text{and} \quad \text{Pe}_L \equiv H_1 L / D_L F \varrho_L (1 - e_G). \quad (3a,b)$$

The rate of mass transfer from gas to liquid can be expressed as

$$\frac{K_L a F H_1}{L} (x^* - x) = \frac{dx}{dw} - \frac{1}{\text{Pe}_L} \frac{d^2x}{dw^2}. \quad (4)$$

Using Henry's law in the form:

$$x^* = y/m \quad (5)$$

we can eliminate in Eq. (4) the equilibrium liquid phase concentration  $x^*$  corresponding to the local gas phase concentration and get

$$\frac{d^2x}{dw^2} - \text{Pe}_L \frac{dx}{dw} - \lambda N_{OG} \text{Pe}_L x + \frac{G}{L} N_{OG} \text{Pe}_L y = 0, \quad (6)$$

where

$$N_{OG} \equiv (K_L a F H_1) / (\lambda L) \quad (7a)$$

and

$$\lambda \equiv m(G/L). \quad (7b)$$

On introducing the new concentrations defined as

$$X = m(x/y_1) \quad \text{and} \quad Y = y/y_1 \quad (8a,b)$$

the Eqs (2) and (6) can be written as follows:

$$\frac{d^2X}{dw^2} - \text{Pe}_L \frac{dX}{dw} + \lambda \frac{\text{Pe}_L}{\text{Pe}_G} \frac{d^2Y}{dw^2} + \lambda \text{Pe}_L \frac{dY}{dw} = 0, \quad (9)$$

$$\frac{d^2X}{dw^2} - \text{Pe}_L \frac{dX}{dw} - \lambda N_{OG} \text{Pe}_L X + \lambda N_{OG} \text{Pe}_L Y = 0. \quad (10)$$

A number of comments have been published on the boundary conditions of such equations. Those originally suggested by Damköhler<sup>6</sup> give physically satisfying results. At the liquid inlet we may employ the relations

$$(X)_{w \rightarrow +0} = X_3 + \frac{1}{\text{Pe}_L} \left( \frac{dX}{dw} \right)_{w \rightarrow +0} \left. \vphantom{(X)_{w \rightarrow +0}} \right\} w = 0. \quad (11a)$$

$$dY/dw = 0 \quad (11b)$$

Similarly at the liquid exit from the column we have

$$dX/dw = 0 \quad \left. \vphantom{dX/dw} \right\} w = 1. \quad (12a)$$

$$(Y)_{w \rightarrow -1} = 1 - \frac{1}{\text{Pe}_G} \left( \frac{dY}{dw} \right)_{w \rightarrow -1} \quad (12b)$$

The inlet concentration jumps are characteristic of the conditions (11a) and (12b) which result from the removal of solute at the inlet of respective phases by turbulent mixing. In the case of piston flow through the column, the value of the Peclet number would diverge and the concentration jump would vanish. At the other extreme of perfect mixing, the Peclet number would vanish, the concentration jump would become maximum and the right hand sides of Eqs (11a) and (12b) become indefinite.

Equations (9) and (10) constitute a set of linear differential equations with constant coefficients. On elimination of either dependent<sup>9-13</sup> variable and its derivatives we obtain the characteristic equation

$$p\{p^3 + (\text{Pe}_G - \text{Pe}_L)p^2 - [\text{Pe}_G\text{Pe}_L + \text{N}_{\text{OG}}(\text{Pe}_G + \lambda \text{Pe}_L)]p + \text{N}_{\text{OG}}\text{Pe}_G\text{Pe}_L(1 - \lambda)\} = 0. \quad (13)$$

The first root is zero ( $p_1 = 0$ ) and the remaining three roots  $p_2, p_3, p_4$  can be found applying the trigonometric solution of the cubic equation<sup>7</sup> within capital brackets in Eq. (13). For all combinations of parameters used in this work the cubic equation had three different real roots. The general solution of equations (9) and (10) is

$$X = c_1 + \sum_{i=2}^4 c_i \exp(p_i w), \quad (14)$$

$$Y = k_1 + \sum_{i=2}^4 k_i \exp(p_i w). \quad (15)$$

The relations between the integration constants in equations (14) and (15) are

found by substituting these integrated solutions into equation (10) of the original system:

$$k_1 = c_1 \quad (16)$$

$$k_i = [p_i(1 - p_i/Pe_L)/\lambda N_{OG} + 1] c_i \quad i = 2, 3, 4. \quad (17)$$

The constants for the desired particular solution are determined with the aid of the boundary conditions (11a,b) and (12a,b):

$$c_1 = X_3 - \sum_{i=2}^4 c_i(1 - p_i/Pe_L), \quad (18a)$$

$$c_2 = (1 - X_3)(K_3L_4 - K_4L_3)/D, \quad (18b)$$

$$c_3 = (1 - X_3)(K_4L_2 - K_2L_4)/D, \quad (18c)$$

$$c_4 = (1 - X_3)(K_2L_3 - K_3L_2)/D, \quad (18d)$$

where

$$D = K_2(L_3M_4 - L_4M_3) + K_3(L_4M_2 - L_2M_4) + K_4(L_2M_3 - L_3M_2), \quad (19)$$

$$K_i = p_i[p_i(1 - p_i/Pe_L)/\lambda N_{OG} + 1], \quad (20)$$

$$L_i = p_i \exp(p_i) \quad i = 2, 3, 4 \quad (21)$$

$$M_i = [\exp(p_i)](1 + p_i/Pe_G)[p_i(1 - p_i/Pe_L)/\lambda N_{OG} + 1] + [(p_i/Pe_L) - 1]. \quad (22)$$

The solutions give gas and liquid concentration profiles within the bubble column. The mass transfer mechanism is dependent on the rate of mass transfer as expressed by the number of overall transfer units  $N_{OG}$ , the degree of gas mixing as expressed by the Peclet number  $Pe_G$ , the degree of liquid mixing as expressed by the Peclet number  $Pe_L$  and the operating conditions as expressed by the ratio  $\lambda$ . The Peclet number can range from zero when a respective phase is completely mixed to infinity when it flows through the column without axial mixing (piston plow). It is apparent that because of physical interaction of gas and liquid streams in the column the mixing intensity of one phase will affect the degree of mixing of the other phase.

## Concentration Profiles

In order to show an effect of mixing of the gas and liquid on the concentration profiles within the column we made systematic calculation of the concentrations from Eqs (13)–(22) for selected values  $\lambda$ ,  $N_{OG}$ ,  $Pe_G$  and  $Pe_L$ . Results for limiting cases of piston flow or perfect mixing are shown in Figs 2 and 3. It was assumed in all computations that the liquid entering the column did not contain any solute, i.e.  $X_3 \equiv X_{w \rightarrow 0} = 0$ . The horizontal lines correspond to perfect mixing in either phase. It is of interest to note in Fig. 3 that the concentration profiles are practically linear when both gas and liquid flows without mixing (piston flow). The limiting size of the inlet concentration jumps given by Eqs (11a) and (12b) is also visualized in Figs 2 and 3. The concentration discontinuity at the inlet disappears in the case of piston flow of either phase. Size of maximum concentration jump on one phase that corresponds to its perfect mixing is further increased if the other phase is unmixed.

With respect to equations (8a) and (8b) which define  $X$  and  $Y$ , the local liquid concentration can be viewed as the equilibrium gas concentration. Then one can take the a distance between the  $X$  and  $Y$  lines as a reduced driving force of mass transfer between the two phases. When comparing the distance between the lines 1 and 3

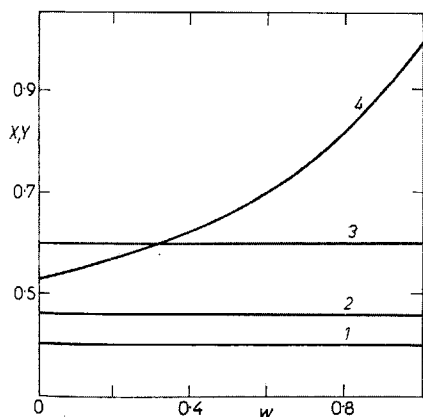


FIG. 2

Concentration Profiles for Perfectly Mixed Liquid ( $Pe_L = 0$ ),  $\lambda = 1$ ,  $N_{OG} = 2$

1 Liquid concentration,  $Pe_G = 0$ ; 2 liquid concentration,  $Pe_G \rightarrow \infty$ ; 3 gas concentration,  $Pe_G = 0$ ; line 4: gas concentration,  $Pe_G \rightarrow \infty$ .

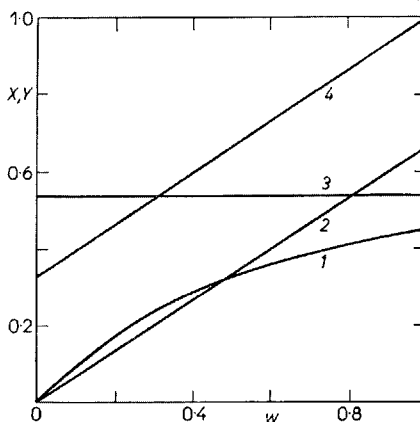


FIG. 3

Concentration Profiles for Piston Flow of Liquid ( $Pe_L \rightarrow \infty$ ),  $\lambda = 1$ ,  $N_{OG} = 2$

1 Liquid concentration,  $Pe_G = 0$ ; 2 liquid concentration,  $Pe_G \rightarrow \infty$ ; 3 gas concentration,  $Pe_G = 0$ ; 4 gas concentration,  $Pe_G \rightarrow \infty$ .

and then between the lines 3 and 4 in Figs 2 and 3 we can see that the driving force is altered by mixing. If both phases are perfectly mixed the driving force is constant along the column. When the liquid phase remains perfectly mixed and the gas phase flows without mixing, the driving force is lowered at the liquid inlet ( $w = 0$ ) and it is considerably increased at the liquid outlet ( $w = 1$ ). In the case of plug flow of both phases the driving force does not practically change with the column length but is considerably greater than in the case of perfect mixing of both phases. If the liquid phase is unmixed and the gas is perfectly mixed the driving force is increased at the liquid inlet and it is decreased at the other end of the column in comparison with the previous case. Numerical values of the driving force for the respective cases are given in Table I.

The mean driving force expressed as the arithmetic mean over the column is the greatest in the case when both the gas and liquid phase flow without mixing. When either phase is perfectly mixed the mean driving force is considerably lowered. Further decrease in driving force is observed if the other phase is also perfectly mixed. For different degrees of mixing in both phases the concentration profiles in the liquid phase are plotted in Figs 4–6. These results show a general region where the concentration profiles are sensitive to the value of the Peclet number. To determine with more accuracy when perfect mixing or piston flow of either phase can be assumed it is also necessary to know to what extent the other phase is mixed.

TABLE I  
Effect of Mixing on Driving Force for  $\lambda = 1$ ,  $N_{OG} = 2$

$Pe_L$	$Pe_G$	Driving force ( $Y - X$ )		Mean driving force
		liquid inlet $w = 0$	liquid outlet $w = 1$	$\frac{\int_0^1 (Y - X) dw}{\int_0^1 (Y - X) dw}$ $Pe_G, Pe_L \rightarrow \infty$
$\infty$	$\infty$	0.33	0.33	1.00
$\infty$	0	0.51	0.07	0.70
0	$\infty$	0.07	0.51	0.70
0	0	0.20	0.20	0.61

### Stage Efficiency

The degree of mixing of both phases is one of the factors influencing the plate efficiency needed for the design of sectionalized bubble columns. For generalized correlations of mass transfer on trays in such columns it is however more suitable to employ quantities such as the number of overall transfer units or the overall mass transfer coefficient than the plate efficiency itself. Among other factors, the relation between these quantities depends on the concentration profiles across a stage and hence on the intensity of mixing of both phases.

The Murphree plate efficiency is defined as

$$E_{MV} \equiv (y_2 - y_1)/(y_2^* - y_1), \quad (23)$$

where

$$y_2^* = mx_2. \quad (24)$$

With respect to the concentrations  $X$  and  $Y$  introduced by Eqs (8a) and (8b) we can rewrite the definition of Murphree plate efficiency as

$$E_{MV} = [1 - (Y)_{w=0}]/[1 - (X)_{w=1}]. \quad (25)$$

Since the systematic calculation of, for example, the number of transfer units from the plate efficiency is tedious, for selected values of  $N_{OG}$ ,  $\lambda$ ,  $Pe_G$  and  $Pe_L$  we calculated the corresponding values of  $E_{MV}$  on a computer. Selected results are shown

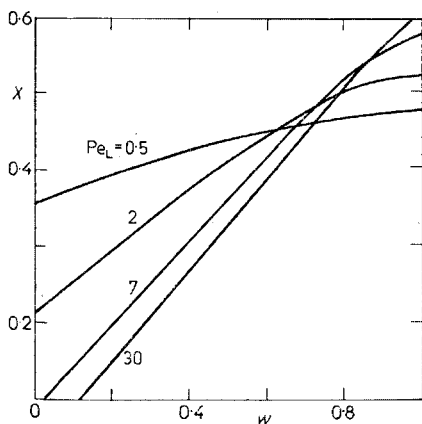


FIG. 4

Concentration Profiles for  $\lambda = 1$ ,  $N_{OG} = 2$  and  $Pe_G = 0.5$

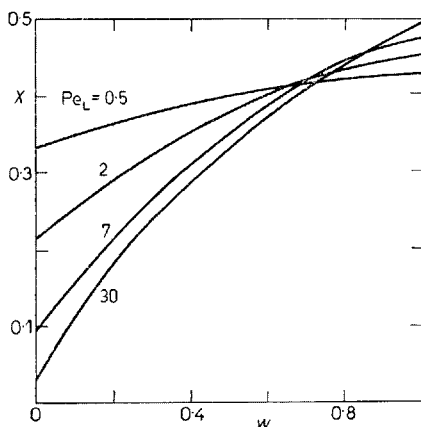


FIG. 5

Concentration Profiles for  $\lambda = 1$ ,  $N_{OG} = 2$  and  $Pe_G = 2.0$



in Figs 7–9. Similarly as in the above computations of the concentrations profiles it was assumed that the liquid entering the stage did not contain any solute. In these calculations the numerical values of the plate efficiency are independent of a value arbitrarily assigned to the inlet liquid concentration as would be expected. The course of the curves in these figures indicates that the plate efficiency is more sensitive to the mixing intensity in the range of lower values of the Peclet number. When comparing

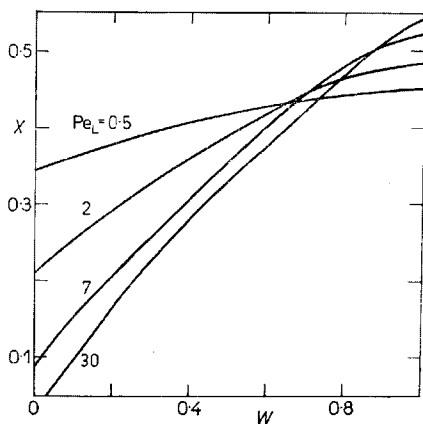


FIG. 6

Concentration Profiles for  $\lambda = 1$ ,  $N_{OG} = 2$  and  $Pe_G = 7$

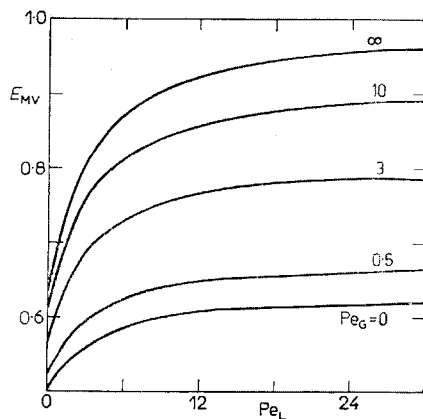


FIG. 7

Dependence of Plate Efficiency on  $Pe_G$  and  $Pe_L$  for  $\lambda = 1$ ,  $N_{OG} = 1$

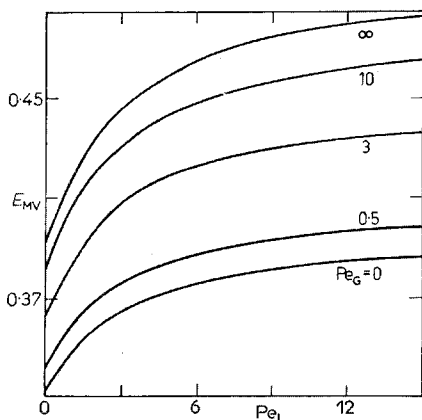


FIG. 8

Dependence of Plate Efficiency on  $Pe_G$  and  $Pe_L$  for  $\lambda = 1$ ,  $N_{OG} = 0.5$

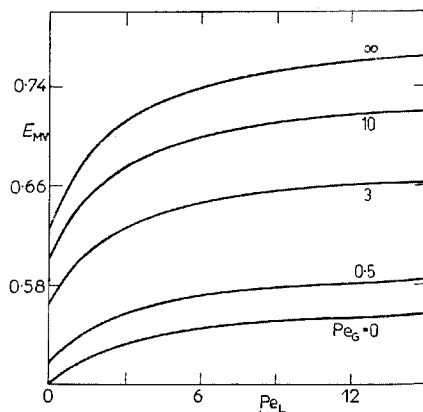


FIG. 9

Dependence of Plate Efficiency on  $Pe_G$  and  $Pe_L$  for  $\lambda = 0.5$ ,  $N_{OG} = 1$

the values plotted in Fig. 7 and 8 one can observe that the effect of mixing is more pronounced when the rate of mass transfer expressed by  $N_{OG}$  is higher. Relative magnitude of mixing effect of either phase is determined by the magnitude of the ratio of equilibrium and operating lines. When this ratio  $\lambda$  is less than unity the plate efficiency is more sensitive to the degree of gas mixing. If  $\lambda$  is greater than unity the liquid mixing has a greater influence on the plate efficiency than mixing of the gas phase. In the case of  $\lambda = 1$  the effect of mixing of the respective phases is of the same magnitude as shown in Table II.

## RESULTS

In our previous work on performance of slotted trays<sup>9</sup> we converted the plate efficiencies to number of overall transfer units taking account of vertical liquid mixing in the foam layer. These values of  $N_{OG}$  were then used in correlations of mass transfer on Turbogrid trays. Measurements of gas residence time in bubble columns reported by Carleton and coworkers<sup>10</sup> and Towell and Ackerman<sup>11</sup> provide the data also for evaluation of an effect of the gas phase mixing. The required values of the Peclet numbers were calculated from Eqs (3a) and (3b). The values of the liquid turbulent diffusivity were taken from the work of Dilman and Senkina<sup>12</sup> and the values of the gas turbulent diffusivity given by Towell and Ackerman<sup>11</sup> were used. These authors measured the turbulent diffusivities under the conditions considerably different from ours (column diameter  $d = 0.150$  m, tray free area 8%, slot width 4 mm, system methanol-water), so that the values of the Peclet numbers presented in Table III, can be considered to be rough approximations only. The plate efficiencies were measured for total reflux over the range of superficial vapor velocities  $0.51-0.67$   $\text{m s}^{-1}$ . The height of foam increased from 0.18 to 0.42 m as the vapour flow rate increased.

TABLE II  
Dependence of Stage Efficiency on Peclet Numbers and Ratio  $\lambda$  for  $N_{OG} = 1$

$Pe_G$	$Pe_L$	$\lambda$					
		0.5		1.0		1.5	
		$E_{MV}$	reduction	$E_{MV}$	reduction	$E_{MV}$	reduction
10	10	0.7135	—	0.8478	—	1.0102	—
0.5	10	0.5794	0.1341	0.6432	0.2046	0.7082	0.3020
10	0.5	0.6228	0.0907	0.6432	0.2046	0.6641	0.3461

TABLE III  
Sample Evaluation of Measurements of Plate Efficiency

$w_G$ $m\ s^{-1}$	$H_1$ m	$E_{MV}$	$D_G$ $m^2\ s^{-1}$	$Pe_G$	$D_L \cdot 10^4$ $m^2\ s^{-1}$	$Pe_L$	$N_{OG}$
0.51	0.18	1.20	0.22	1.06	5	0.55	3.80
0.52	0.21	1.19	0.22	1.27	5	0.68	3.75
0.55	0.24	1.17	0.22	1.53	5	0.86	3.15
0.60	0.29	1.20	0.25	1.74	5	1.09	3.22
0.62	0.34	1.21	0.25	2.11	5	1.34	3.01
0.67	0.42	1.14	0.28	2.51	6	1.43	2.89

As can be seen from Table III the intensity of axial gas and liquid mixing decreases with increasing foam height. The values of the Peclet number for the gas phase are higher than those for the liquid phase by a factor of about two.

Although the operating conditions for bubble columns are somewhat different from those we employed in our experiments it can be seen that a more exact analysis of mass transfer in such columns would require accurate measurements of turbulent diffusivity in both the gas and the liquid. A large effect of the column diameter on the turbulent diffusivity should be also considered.

#### LIST OF SYMBOLS

$c_i$	integration constants given by Eqs (18a)–(18d)
$D_G$	turbulent diffusivity in the gas phase ( $m^2\ s^{-1}$ )
$D_L$	turbulent diffusivity in the liquid phase ( $m^2\ s^{-1}$ )
$e_G$	volumetric fraction of gas
$E_{MV}$	vapour Murphree plate efficiency (Eq. (23))
$F$	cross section of the column ( $m^2$ )
$G$	gas flow rate ( $mol\ s^{-1}$ )
$H$	length of flow path in the column from inlet (m)
$H_1$	height of aerated liquid (m)
$k_i$	integration constants given by Eqs (16), (17), (18a)–(18d)
$K_{La}$	overall gas mass transfer coefficient ( $mol\ m^{-3}\ s^{-1}$ )
$L$	liquid flow rate ( $mol\ s^{-1}$ )
$m$	Henry's law constant
$N_{OG}$	number of overall gas phase transfer units (Eq. (7))
$Pe_G$	Peclet number for the gas phase (Eq. (3a))
$Pe_L$	Peclet number for the liquid phase (Eq. (3b))
$w = H/H_1$	relative flow path length through aerated liquid
$w_G$	linear superficial gas velocity ( $m\ s^{-1}$ )
$x$	liquid mole fraction of solute

$x_2$	exit liquid mole fraction of solute
$x_3$	inlet liquid mole fraction of solute
$x^*$	liquid mole fraction of solute in equilibrium with gas, $y$ (Eq. (5))
$X = mx/y_1$	concentration of solute in liquid
$y$	gas mole fraction of solute
$y_1$	inlet gas mole fraction of solute
$y_2$	exit gas mole fraction of solute
$Y = y/y_1$	concentration of solute in gas
$\lambda \equiv mG/L$	ratio of slope of equilibrium and operating line
$\rho_G$	molar density of gas ( $\text{mol m}^{-3}$ )
$\rho_L$	molar density of liquid ( $\text{mol m}^{-3}$ )

## REFERENCES

1. Fair J. R.: Chem. Eng. (New York) 67, July 3 (1967).
2. Fair J. R.: Chem. Eng. (New York) 207, July 17 (1967).
3. Mashelkar R. A.: Brit. Chem. Eng. 15, 1297 (1970).
4. Hartman M., Standart G.: This Journal 32, 1166 (1967).
5. Pavlica R. T., Olson J. H.: Ind. Eng. Chem. 62, 45 (1970).
6. Damköhler G.: Z. Elektrochem. 43, 1 (1937).
7. Korn A. G., Korn T. M.: *Mathematical Handbook for Scientists and Engineers*. McGraw-Hill, New York 1968.
8. Mhaskar R. D.: Chem. Eng. Sci. 29, 897 (1974).
9. Hartman M., Grigar K., Standart G.: Chem. Process (London) 48, (8), 76 (1967).
10. Carleton A. J., Flain R. J., Rennie J., Valentin F. H. H.: Chem. Eng. Sci. 22, 1839 (1967).
11. Towell G. D., Ackerman G. H.: Proceedings of the Fifth European/Second International Symposium on Chemical Reaction Engineering, B 3-1, Amsterdam 1972.
12. Dilman V. V., Senkina E. V.: Khim. Tekhnol. Topl. Masel No 8, 46 (1964).
13. Wylie C. R. jr: *Advanced Engineering Mathematics*. McGraw-Hill, New York 1966.