Applicability of Axial Dispersion Model to Analyze Mass Transfer Measurements in Bubble Columns

The applicability of axial dispersion model (*ADM*) for the measurement of the gas-liquid mass transfer coefficient (k_La) in bubble columns is discussed. It is shown that the misinterpretation of the concentration jump near the column inlet can lead to wrong conclusions regarding the mass transfer rates. It is further illustrated that some reported k_La dependencies on the superficial liquid velocity and the axial distance are resulted from the use of an incorrect model and can be rectified by an application of *ADM*. Finally, it is shown that *ADM* can be effectively used to calculate k_La for: (a) highly viscous liquids, (b) in the presence of chemical reaction, (c) slurry reactors, and (d) the columns with different liquid inlet locations.

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SCOPE

The axial dispersion model (*ADM*) has been extensively used to analyze the gas-liquid mass transfer characteristics of bubble columns. Recently, Alvarez-Cuenca and coworkers (Alvarez-Cuenca et al., 1980, 1980a; Alvarez-Cuenca and Nerenberg, 1981), and Herbrechtsmeier et al. (1981) have concluded that *ADM* is not applicable under certain operating conditions. Owing to the growing interest in this mass transfer phenomenon, it seems timely: first, to clarify the contradictory findings; and second, to point out once again that *ADM* is well suited to determine the k_La data in bubble columns from the measured concentration profiles. This is achieved:

(1) By showing that the evaluation of k_La is not strongly affected by the liquid-phase dispersion coefficient. Nevertheless, dispersion must not be neglected.

(2) By critically analyzing the work of Alvarez-Cuenca and Nerenberg (1981), and showing that under physically reasonable assumptions, their data can be evaluated with the help of *ADM.*

(3) By demonstrating that the use of incorrect model leads to dependencies of k_La on the liquid velocity and the dispersion height, which are not observed if a more appropriate model, i.e., the *ADM*, is applied.

(4) By presenting new and updated measurements for oxygen mass transfer in various bubble columns under different set of operating conditions.

CONCLUSIONS AND SIGNIFICANCE

The applicability of axial dispersion model (*ADM*) is tested over a wide range of operating conditions. Following conclusions can be drawn from the results:

- When applying *ADM*, it should be remembered that the dispersion is a macroscopic phenomenon. Therefore, dispersed flow may not be fully developed at the system boundaries; for instance, in the close vicinity of the gas sparger.
- In partially backmixed reactors, one must be aware of the concentration jump at the reactor inlet. This jump is a result of backmixing and must not be interpreted as an enhanced mass transfer rate.
- Dependencies of k_La on the liquid velocity and the column height are an outcome of the application of wrong model, *PFM.* If an appropriate model, *ADM*, is used, such dependencies disappear.
- For k_La determinations in bubble column reactors, measurements of steady-state oxygen profiles are strongly recommended. Evaluation of such profiles on the basis of the *ADM* gives consistent and conclusive k_La data, which can be correlated by simple relations, and in addition, are independent of the column scale and the flow direction (countercurrent flow, cocurrent upflow and downflow).
- The *ADM* can be successfully applied to evaluate mass transfer data in aerated, highly viscous media where plug flow prevails, and in slurry reactors where nonuniform particle distribution exists.

INTRODUCTION

The volumetric mass transfer coefficient k_La in bubble columns is normally evaluated based on one of the following assumptions for the flow pattern of the liquid phase: (a) complete mixing (continuous stirred-tank reactor model); (b) plug flow (plug-flow model); and (c) assuming dispersed plug flow in the liquid phase (axial dispersion model). Applicability of the continuous stirred-tank reactor model (*CSTRM*) is only restricted to large diameter columns, say $D_c > 0.3$ m, or small length to diameter ratios (between 1 and 3).

Among others, the plug-flow model (*PFM*) has been used by

TABLE 1. BUBBLE COLUMNS USED IN O_2 MASS TRANSFER STUDIES

Bubble Column	Diameter m	Height m	Gas Sparger
BC I	0.2	7.2	Cross with 56 orifices of 1-mm diameter
BC II	0.15	4.4	Sintered plate with mean pore diameter of 150 μm
BC III	0.2	2.5	Same as BC I
BC IV	0.14	2.7	Various interchangeable sintered and perforated plates

Voyer and Miller (1968) and Alvarez-Cuenca et al. (1980). However, the flow pattern in bubble columns is often close to mixed flow, but not completely backmixed. Therefore, the assumption of a partially backmixed liquid phase is more appropriate. Especially in tall and relatively small diameter bubble columns the k_La values should be determined with the help of concentration profiles measured along the length of the column for cocurrent or countercurrent flow and evaluated by means of the axial dispersion model (*ADM*). Chang (1970), Reuß (1970), Deckwer et al. (1974, 1981), Fan et al. (1975), Schügerl et al. (1977), Shioya and Dunn (1978), Ying et al. (1980), and Mangartz and Pilhofer (1981) have reported the mass transfer coefficients based on the axial dispersion model (*ADM*).

Recently, Alvarez-Cuenca and coworkers (Alvarez-Cuenca et al., 1980, 1980a; Alvarez-Cuenca and Nerenberg, 1981) have published a series of articles reporting the mass transfer rates in a bubble column at high gas and liquid throughputs. While these authors should be commended for the extensive and methodical presentations of oxygen concentration contour diagrams, it should be pointed out that their evaluations and conclusions are, at least partly misleading regarding the applicability of *ADM*. Alvarez-Cuenca et al. (1980) used a *PFM* to evaluate k_La data. In another paper, Alvarez-Cuenca and Nerenberg (1981) proposed a two-zone model (T-ZM) which is a modified *PFM*, to explain the measured oxygen concentration profiles. In a recent study of a downflow bubble column, Herbrechtsmeier et al. (1981) also presented a few measured axial oxygen concentration profiles. These authors reported again that their profiles could not be described with the help of the *ADM*. When using the *ADM* for a point-wise description of the measured oxygen concentrations, Herbrechtsmeier et al. (1981) found a strong spatial dependency of k_La.

In this paper the data of Alvarez-Cuenca et al. (1980) and Herbrechtsmeier et al. (1981) are reanalyzed using *ADM* to shed further light on the above contradictory reports. Also, the versatility of *ADM* is demonstrated with the help of additional data under different operating conditions. The geometrical sizes of the columns and the gas spargers used are listed in Table 1. The experimental techniques and the method of k_La determination are described in detail elsewhere (Deckwer et al., 1974, 1981; Burchart, 1976; Hallensleben, 1980; Nguyen-tien, 1981).

EFFECT OF LIQUID-PHASE DISPERSION ON EVALUATION OF k_La

The differential equation of the *ADM* for the liquid phase can be written as

$$\frac{1}{Pe_L}\frac{d^2c_L}{dz^2} + a'\frac{dc_L}{dz} + St_L(c_L^* - c_L) = 0 \tag{1}$$

where

$$Pe_L = \frac{u_L L}{\epsilon_L D_L} \tag{2}$$

$$St_L = k_La\frac{L}{u_L} \tag{3}$$

a' denotes the flow direction of the phases ($a' = -1$ for cocurrent flow, $a' = 1$ for countercurrent flow). Equation 1 is subjected to Danckwerts' boundary conditions; for cocurrent flow

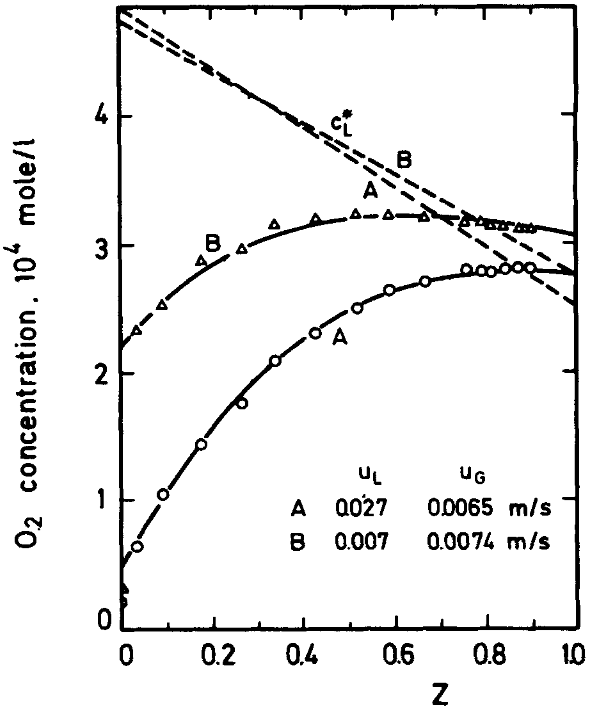

Figure 1. O_2 concentration profiles in BC I.

$$c_{Li} = C_L|_{z=0} - \frac{1}{Pe_L}\frac{dc_L}{dz}\bigg|_{z=0} \tag{4}$$

$$\frac{dc_L}{dz}\bigg|_{z=1} = 0 \tag{5}$$

for countercurrent flow

$$\frac{dc_L}{dz}\bigg|_{z=0} = 0 \tag{6}$$

$$c_{Li} = c_L|_{z=1} + \frac{1}{Pe_L}\frac{dc_L}{dz}\bigg|_{z=1} \tag{7}$$

The equilibrium concentration c_L^* is given by Henry's law

$$c_L^* = Px/H \tag{8}$$

In the case of oxygen mass transfer, it can be assumed that the O_2 mole fraction x is approximately constant along the column height as the O_2 solubility is small. However, in tall bubble columns one has to consider that the total pressure P depends on z due to the hydrostatic head.

The analytical solution of Eq. 1 is given elsewhere (Langemann, 1968; Deckwer et al., 1974). Two dimensionless groups—the Peclet number Pe_L and the Stanton number St_L—are involved in Eq. 1. If the Peclet number can be determined by independent means, in principle, the Stanton number can be calculated from only one single measurement of the oxygen liquid-phase concentration. However, at gas velocities prevailing in industrial application and in tall bubble columns equipped with efficient gas spargers, this procedure does not apply as the liquid phase at the column top may be supersaturated. Figure 1 presents two measured concentration profiles for bubble column (*BC*) I. Although the gas flow of these measurements is small, it is seen that the liquid becomes supersaturated at the column top. From the measured concentration profile of oxygen in the liquid phase, both parameters involved in Eq. 1 can be obtained explicitly. To check the sensitivity of k_La determination against variations of Pe_L, the square sum of the deviations of the measured data (run B in Figure 1) from the model predictions is plotted vs. St_L for different values of Pe_L in Figure 2. This figure reveals a remarkable result that the location of the minimum is rather insensitive to changes in Pe_L. If Pe_L varies from 0.5 to 10 the corresponding Stanton number varies merely from 3.8 to 4.4.

The above observation implies that the two-parameter optimi-

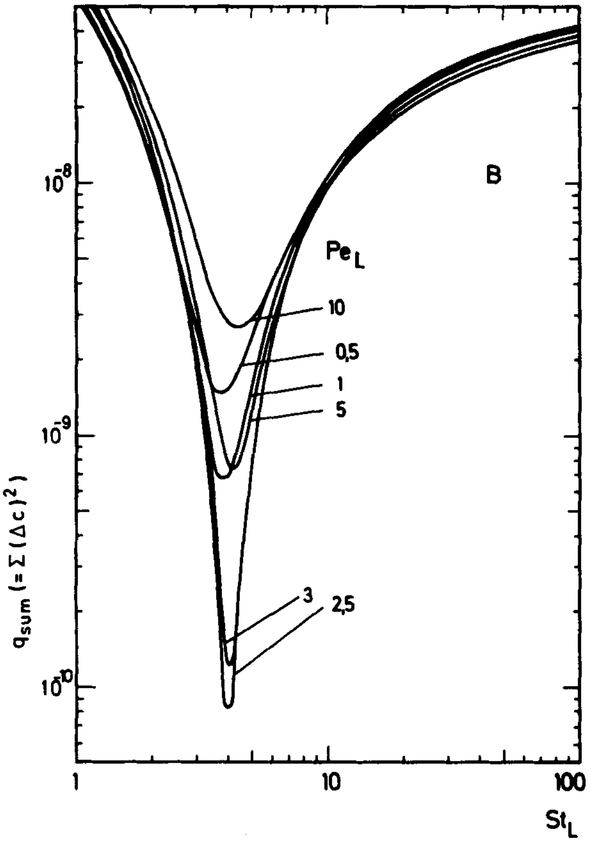

Figure 2. Square sum deviation as function of St_L for various Pe_L.

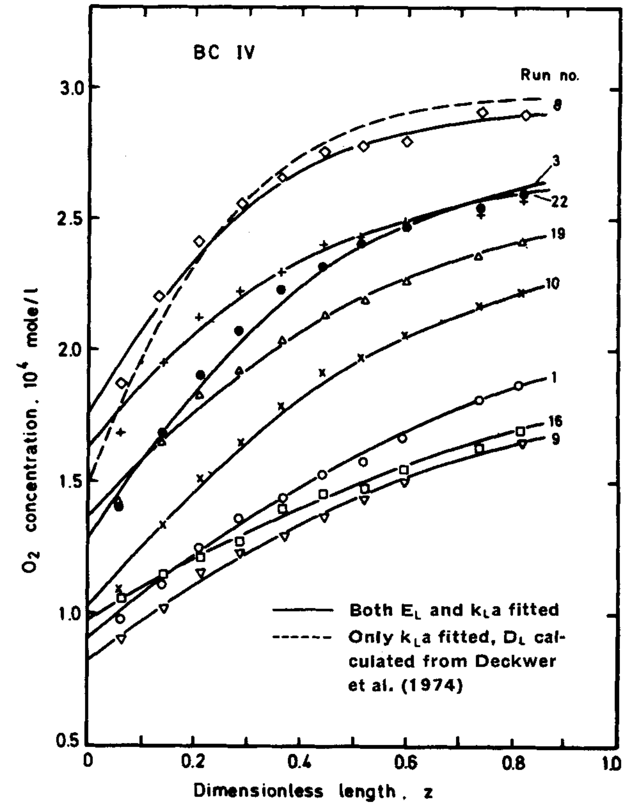

Figure 3. *ADM* description of O_2 profiles in BC IV at cocurrent flow (perforated plate with 1-mm bores).

zation problem can often be reduced to a one-parameter problem by estimating the value of the Peclet number from correlations given in the literature (Deckwer et al., 1974; Joshi, 1980).

Some O_2 profiles measured in BC IV are shown in Figure 3. They can be excellently described with the *ADM* when both D_L and k_La are varied to fit the data. Table 2 gives the operating conditions for the runs shown in Figure 3 and also the D_L and k_La values. Only at low gas velocities and $u_L = 0.054$ m/s, the optimized D_L and the D_L values calculated from the correlation of Deckwer et al. (1974) show reasonable agreement. This correlation was established from measurements at low liquid velocities ($u_L \leq 0.03$ m/s) and does not consider an effect of u_L. There might be such an effect at higher liquid flow rates as indicated by Joshi (1980) and Kara et al. (1981). Although the predicted and fitted D_L values differ considerably, the k_La data obtained from the one- and two-parameter models differ only slightly (less than 10%; Table 2). This verifies the conclusions derived from Figure 2 that the concentration profile fitting is remarkably sensitive to variations of k_La but rather insensitive to changes in D_L.

Despite the fact that k_La determinations on the basis of the *ADM* are influenced only slightly by the numerical value of the Peclet number, it is not desirable to eliminate this group from Eq. 1 and to make use of the *PFM*, as this results in the misinterpretation of the concentration jump at the column inlet. Alvarez-Cuenca et al. (1980) and Alvarez-Cuenca and Nerenberg (1981) interpreted this concentration jump near the gas sparger as being due to high mass transfer rates. However, the concentration jump is merely a result of the partial backmixing in the liquid phase and this is clearly indicated by Danckwerts' boundary conditions.

ANALYSIS OF WORK OF ALVAREZ-CUENCA AND COWORKERS

The measurements of these authors (Alvarez-Cuenca, 1980, 1980a; Alvarez-Cuenca and Nerenberg, 1981) were carried out in a rectangular column of special sizes (0.025 m thick, 0.66 m wide and 2.5 m tall). First, it should be admitted that their column cannot be regarded as a conventional bubble column. Their column was aerated with the help of four equally spaced nozzles. The authors have presented a schematic diagram of sampling point distributions (Alvarez-Cuenca and Nerenberg, 1981). It is clearly seen that the

TABLE 2. VOLUMETRIC MASS TRANSFER COEFFICIENT k_La FROM O_2 CONCENTRATION PROFILES MEASURED IN BC IV

Run No.	u_G m/s	u_L m/s	$10^5 c_{Li}$ mol/L	D_L calc m²/s	k_La^{+} fitted L/s	D_L^{++} fitted m²/s	k_La^{++} fitted L/s
1	0.014	0.054	7.81	0.0118	0.0136	0.0109	0.0137
3	0.039	0.054	7.77	0.0175	0.0403	0.0223	0.0397
8	0.124	0.054	6.25	0.0231	0.0858	0.0487	0.0814
9	0.016	0.077	7.19	0.0125	0.0165	0.0195	0.0160
10	0.032	0.077	7.81	0.0154	0.0329	0.0230	0.0321
11	0.049	0.077	8.59	0.0175	0.0431	0.0299	0.0416
13	0.082	0.075	7.03	0.0204	0.0682	0.0338	0.0663
16	0.016	0.108	9.06	0.0125	0.0204	0.0208	0.0198
19	0.066	0.108	9.38	0.0191	0.0617	0.0571	0.0575
22	0.166	0.108	10.16	0.0226	0.0887	0.0886	0.0812

[+] k_La from profile fitting with D_L calculated by Deckwer et al. (1974).

[++] Both k_La and D_L optimized by profile fitting.

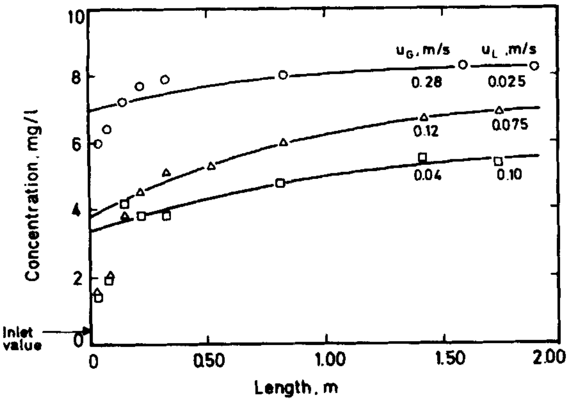

Figure 4. *ADM* fit of O_2 concentration profiles reported by Alvarez-Cuenca and Nerenberg (1981).

first sampling point is in the same line of nozzles while the next two points are in the close vicinity (about 0.15 m).

While applying the *ADM* one has to realize that the dispersion in bubble columns is a macroscopic phenomenon. The sizes of the single steps of mass propagation by dispersion are in the range of bubble sizes or even larger. Hence, fully established dispersive flow can only be expected after a number of single events has occurred, i.e., after the gas phase has travelled a certain distance. This is supported by experimental evidence as the oxygen concentration measured in the close vicinity of the gas sparger reveals extreme fluctuations. Therefore, the measurements in this entrance region should be treated with great care, as misleading conclusions can be reached.

Figure 4 shows some measured concentration data points of Alvarez-Cuenca and Nerenberg (1981). The authors could not correlate these data using *ADM* because they tried to include in their analysis the data points close to the gas distributor where dispersive flow is not yet fully developed. In addition, Alvarez-Cuenca and Nerenberg tried to evaluate the inlet oxygen concentration value with the help of profile within the column. This is not possible due to the concentration jump, as implied by the boundary condition. Figure 4 also shows that all data points except the first one or two can be fairly well represented by the *ADM*. Only one single value of k_La is needed to describe each set of data while the two-zone model description of Alvarez-Cuenca and Nerenberg yielded two k_La values which differ by one or more orders of magnitude. Table 3 shows the value of k_La and the dispersion coefficients evaluated by means of the *ADM*. The values of the volumetric mass transfer coefficients are in the expected range but the dispersion coefficients are abnormally high, probably as a result of the different shape of the bubble column. Stiegel and Shah (1977) also observed enlarged dispersion coefficients in rectangular bubble columns. Moreover, the high dispersion coefficients obtained by fitting the profiles with the *ADM* should not result in wrong predictions of mass transfer coefficients as it is already shown that St_L is rather insensitive to Pe_L.

SPATIAL DEPENDENCY OF k_La AND INFLUENCE OF LIQUID VELOCITY

The application of an incorrect model may show dependencies which in reality do not exist. k_La values in bubble columns were found to be spatially dependent and additionally influenced by the liquid velocity by Voyer and Miller (1968), Alvarez-Cuenca et al. (1980, 1980a), and Alvarez-Cuenca and Nerenberg (1981). In Figure 3 axial oxygen concentration profiles are shown for BC IV. It has already been pointed out that these profiles can be excellently described on the basis of the *ADM* with one single value of k_La. If the same data are analyzed with the help of the *PFM* as described by Alvarez-Cuenca et al. (1980), the results shown in Figure 5 (for runs 11 and 19 of Table 2) are obtained. In this case the volumetric mass coefficient shows a spatial dependency. This figure is clearly misleading since it gives the impression that high mass transfer rates are observed near the gas distributor. However, this is also the result of the application of an incorrect model, i.e., the *PFM*, where the occurrence of the concentration jump at the inlet of the axially dispersed reactor is misinterpreted as an increase in the mass transfer rate. Therefore, the finding of Alvarez-Cuenca et al. (1980) that "65 to 95% of the total oxygen transfer takes place within the grid region" (i.e., the zone close to the sparger) is incorrect. Furthermore, the conclusions of these authors concerning the inadequacy of the *ADM* for the mass transfer in bubble column are rather misleading.

TABLE 3. VALUES OF MASS TRANSFER AND DISPERSION COEFFICIENTS EVALUATED WITH *ADM* FROM PROFILES REPORTED BY ALVAREZ-CUENCA AND NERENBERG (1981)

u_G m/s	u_L m/s	k_La L/s	$D_L\epsilon_L$ m^2/s
0.12	0.075	0.0766	0.081
0.28	0.025	0.0764	0.098
0.04	0.100	0.0594	0.154

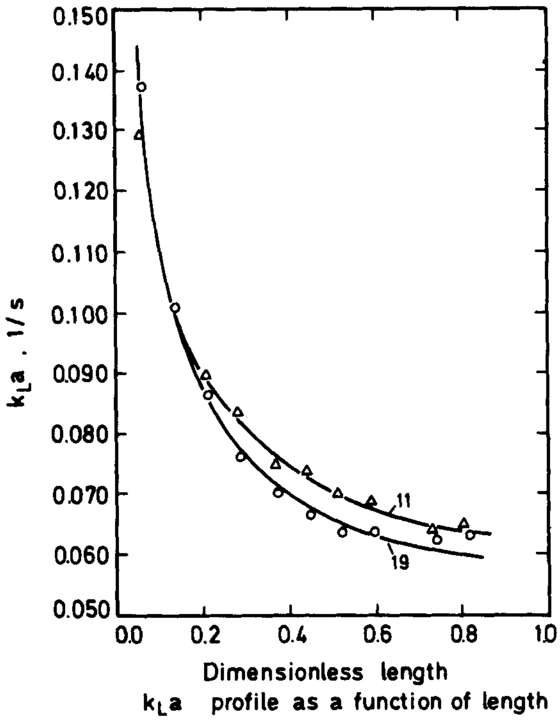

Figure 5. Profiles (Run No. 11 and 19 of Table 2; also Figure 3) evaluated for k_La with the help of *PFM* as used by Alvarez-Cuenca et al. (1980).

Figure 6 illustrates some k_La values as functions of gas and liquid velocities for various bubble columns. The k_La values reported here were obtained from the measurements in BC I (both with cocurrent and countercurrent flow), III and IV (equipped with a perforated plate sparger) and with the use of *ADM*. In BC I and BC II, u_L was varied from 0.01 to 0.03 m/s while in BC IV u_L was raised up to 0.12 m/s. These results show that k_La is relatively independent of the liquid velocity and can be correlated to the gas velocity by the expression

$$k_La = b\,u_G^n$$

where $n = 0.82$ and $b = 0.467$. It has been pointed out elsewhere (Shah et al., 1982) that only b is strongly affected by liquid-phase properties and the type of sparger while n is approximately constant. The data shown in Figure 6 are also in excellent agreement with the predictions of the correlation of Akita and Yoshida (1973) which is generally recommended for k_La estimations in bubble columns with less effective gas spargers (Deckwer, 1980; Shah et al., 1982).

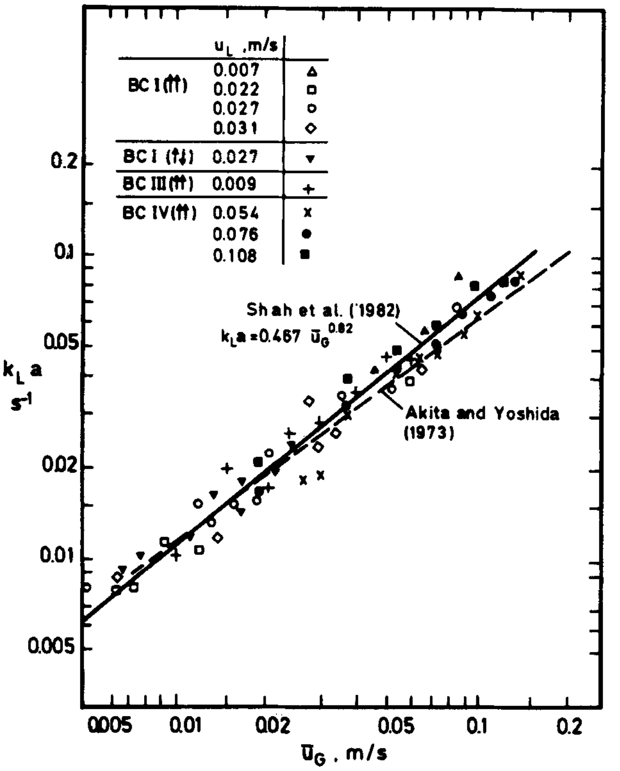

Figure 6. k_La data (using *ADM* fit) from profiles measured in BC I, III and IV for liquid velocities ranging from 0.01 to 0.12 m/s.

In contrast to our findings, both Voyer and Miller (1968) and Alvarez-Cuenca et al. (1980) (who have analyzed their mass transfer data with the help of the *PFM*), reported a pronounced dependency of k_La on the liquid velocity. As shown in Figure 7, an analysis of some profiles measured in BC IV on the basis of *PFM* indicated a clear dependency of k_La on u_L. This figure incorrectly indicates that the mass transfer rate increases with increasing liquid velocity, a result which can be eliminated with the use of the correct model.

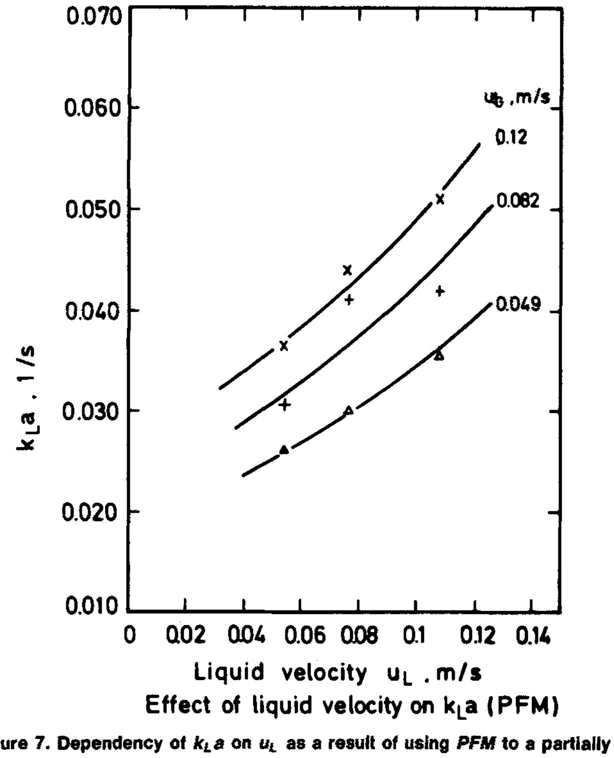

Figure 7. Dependency of k_La on u_L as a result of using *PFM* to a partially backmixed contactor.

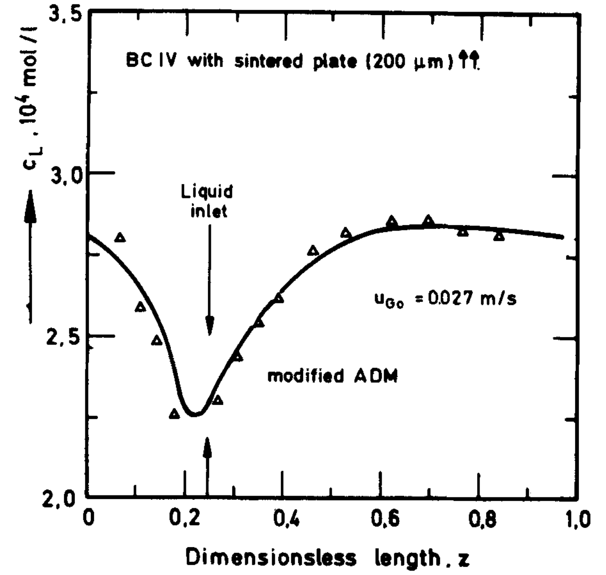

Figure 8. Measured O_2 profile and *ADM* description in BC IV with liquid inlet 0.68 m above sintered plate sparger (Hallensleben, 1980).

It should be pointed out that k_La need not be constant along the column length. In fact, the formation of bubbles at the gas sparger and the resulting enhanced turbulence may cause higher mass transfer rates. However, this was not observed for O_2 mass transfer in water and aqueous solutions of electrolytes if the measured profiles were evaluated with the help of the *ADM* (Deckwer et al., 1974, 1981; Schügerl et al., 1977). Only in fermentation media a decrease in the oxygen uptake rate was observed along the column (Buchholz, 1979; Luttmann, 1980). This was attributed to strong coalescence. By introducing an appropriate coalescence function, the O_2 concentration profile could again be well described by the *ADM*. For CO_2 mass transfer, too, increased k_La values in the vicinity of the sparger were found (Deckwer et al., 1978). However, in this case the CO_2 profiles measured in the gas and the liquid phases could be well described by a modified *ADM*.

ADDITIONAL EVIDENCES OF *ADM* APPLICABILITY

Variation of Liquid-Phase Inlet Location

In industrial bubble columns, the liquid phase is not necessarily introduced at the top or bottom of the column. It was therefore

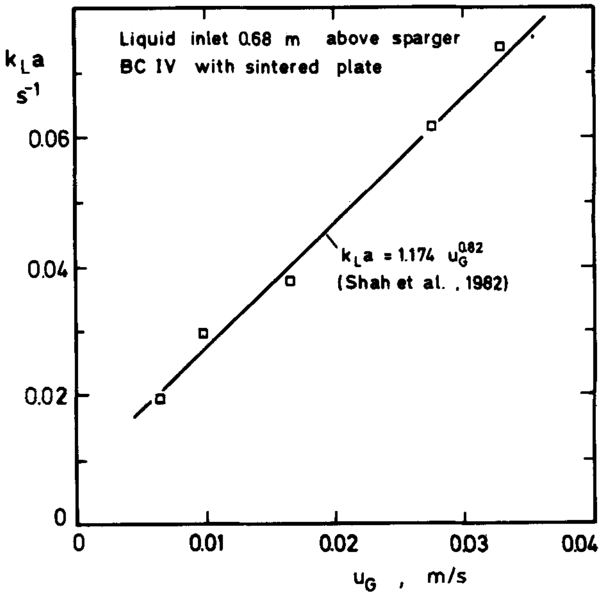

Figure 9. k_La vs. u_G for bubble columns equipped with sintered plate sparger (Hallensleben, 1980).

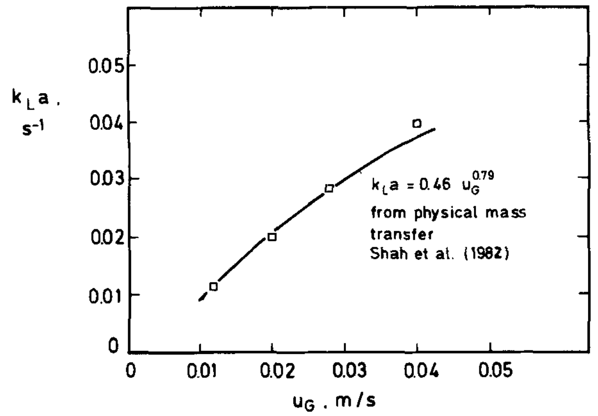

Figure 10. k_La data from CO_2 absorption in carbonate buffer (Quicker et al., 1982) compared with k_La data from physical measurements analyzed with *ADM*.

interesting to check whether the *ADM* applies if the liquid phase is introduced at an arbitrary position along the column. Such measurements were carried out in BC IV equipped with a sintered plate (mean pore size 200 μm). A measured profile with the liquid inlet located 0.68 m above the sparger is shown in Figure 8. As shown, a modified *ADM* fits the measured profile well for k_La = 0.062 s^{-1} (Hallensleben, 1980). In addition, the k_La values obtained for the cases shown in Figure 8 and 9 are in good agreement with the established correlation for various bubble columns with sintered plates as gas distributors and operated in the normal manner, i.e., with a liquid inlet at the column bottom (Shah et al., 1982).

Oxygen Transfer into Highly Viscous Media

Highly viscous non-Newtonian media, such as solutions of carbomethoxycellulose (CMC) have been successfully used to simulate the properties of fermentation broths for antibiotics production (Deckwer et al., 1982). With such high viscous solutions bubbly flow can be observed only at low gas velocities, say $u_G \leq 0.01$ m/s. For $u_G \geq 0.02$ m/s, slug flow prevails and the sizes of the slugs approach the column diameter (0.14 m) with increasing height and gas velocities. However, it was shown (Deckwer et al., 1982) that for fully established slug flow, the *ADM* reasonably describes the measured O_2 profiles. The evaluated k_La data follow a simple correlation.

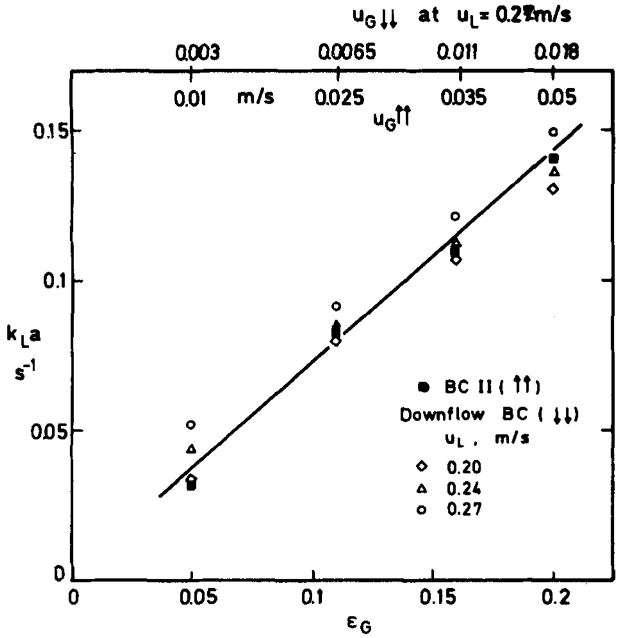

Figure 11. Comparison of k_La data from cocurrent downflow (sulfite oxidation) and cocurrent upflow (physical O_2 transfer) bubble columns (Herbrechtsmeier and Steier, 1978).

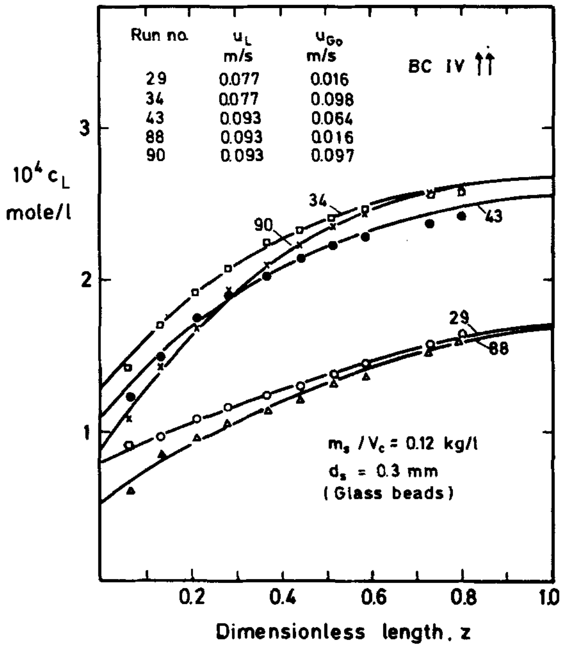

Figure 12. Description of O_2 concentration profiles with *ADM* in presence of glass beads.

Comparison with Chemical Methods

Volumetric mass transfer coefficients k_La can also be determined in the presence of chemical reactions. In particular, if the absorption-reaction process takes place in the diffusional regime (i.e., fast reaction) or the transition regime from slow to fast reaction; the bulk concentration of the solute gas approaches zero. Under this condition, the mixing behavior of the liquid phase does not at all affect the evaluation of k_La data. Quicker et al. (1982) studied the arsenite catalyzed CO_2 absorption in buffer solution. From their measurements in a bubble column of 0.095-m diameter and equipped with a perforated plate sparger, it was possible to determine k_La values by preparing Danckwerts' plots. The four data points reported by Quicker et al. (1982) are given in Figure 10. They are in reasonable agreement with the correlation proposed by Shah et al. (1982) from physical mass transfer data in electrolyte solutions for various bubble columns with multiorifice sparger, i.e., BC I, III and IV.

Mass transfer in a cocurrent downflow bubble column was studied by Herbrechtsmeier and Steiner (1978) using the sulfite oxidation method. A catalyst concentration was used such that the operation in the diffusional reaction regime with a bulk liquid O_2 concentration of about zero was guaranteed. The k_La values determined with porous plates spargers and for three different liquid velocities are plotted as a function of the gas holdup in Figure 11. The data compared well with those of Deckwer et al. (1974) for bubble columns, which were evaluated with the help of *ADM*. It should be noted from Figure 11 that the gas velocities required to obtain a certain gas holdup value are quite different for cocurrent upflow and downflow bubble columns (as indicated by the upper scale in Figure 11) and therefore as proposed by Herbrechtsmeier and Steiner (1978) only the holdup values are a reasonable basis for comparing k_La data in these two systems.

Presence of Solids

To demonstrate the applicability of the *ADM* for evaluating mass transfer data for slurry columns, some preliminary results are presented here. The measurements were carried out in BC IV with cocurrent upflow of gas and liquid. The solid particles (glass beads with a mean diameter of 0.3 mm) batchwise were suspended in a

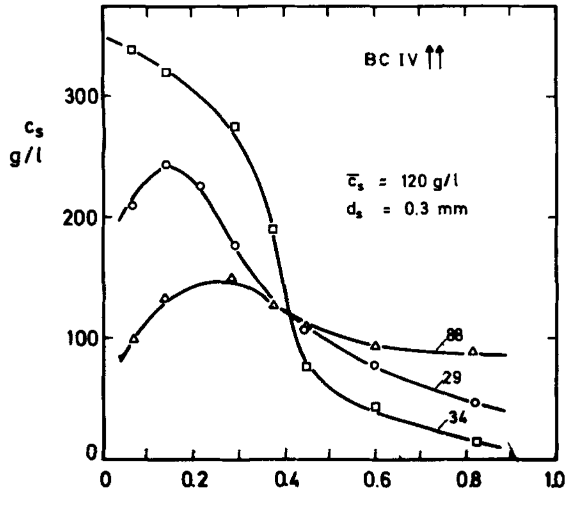

Figure 13. Solid concentration as function of column height.

manner; their removal from the column being prevented by introducing a grid at the column top. Some measured O_2 concentration profiles in the liquid phase are shown in Figure 12. They refer to a mean solid concentration of approximately 0.12 kg per 1 reactor volume (i.e., 5 kg in 41.6 L). The measured data points can be well described by the closed solution of the *ADM*, a k_La value being constant along the entire column length. This result is surprising as the distribution of the solids along the column height can be extremely nonuniform. Typical solid concentration profiles determined simultaneously with the liquid phase O_2 profiles are given in Figure 13. It shows that particularly for high gas velocities the distribution is highly nonuniform due to particle settling (run 34). It is not yet possible to explain the solid concentration profiles quantitatively; however, in spite of this nonuniformity of the solids distribution, no difficulties were encountered in describing the liquid-phase O_2 concentrations with the help of *ADM*.

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NOTATION

ADM = axial dispersion model
c_L = concentration of oxygen in liquid phase, mol/L
c_L^* = equilibrium concentration of oxygen in liquid phase, mol/L
c_{Li} = concentration of oxygen in liquid phase at inlet, mol/L
c_s = solids concentration, g/L
CSTRM = continuous stirred tank reactor model
D_c = column diameter, m
D_L = liquid phase axial dispersion coefficient, m^2/s
d_s = average solid particles diameter, mm
H = Henry's constant, atm·L/mol
k_La = liquid side mass transfer coefficient, s^{-1}
L = length of the column, m
P = pressure, atm
Pe_L = liquid phase Peclet number, defined by Eq. 2
PFM = plug flow model
St_l = liquid phase Stanton number, defined by Eq. 3
u_G = superficial gas velocity, m/s
u_L = superficial liquid velocity, m/s
x = mole fraction of oxygen in gas phase
z = dimensionless axial coordinate

Greek Letters

ϵ_L = liquid-phase holdup

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Numerical Simulation of a Fixed-Bed Adsorption Column by the Method of Orthogonal Collocation

(This is an R&D Note)

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The orthogonal collocation method of solving partial differential equations was developed more than a decade ago, largely by Villadsen (1970) and Finlayson (1972) The method is particularly useful for the solution of boundary value problems and, for a given accuracy of solution, has often been found to require less computer time than standard finite difference methods. Although the method has been previously applied to the simulation of fixed-bed reactors (*e.g., Hansen*, 1971; Karanth and Hughes, 1974) and recently to the simulation of an adsorption column (Liapis and Rippin, 1978; Liapis and Litchfield, 1980), it has not yet become widely adopted as a standard technique. To illustrate the application of the method to the prediction of the transient response of an adsorption column, we have solved a complicated linear problem to which an exact analytic solution has recently been derived (Rasmuson and Neretnieks, 1980). A similar nonlinear problem could be solved in exactly the same way, and the property of linearity is of no special advantage to the collocation method. However, the availability of an exact solution provides a convenient check on the accuracy of the collocation solution.

MATHEMATICAL MODEL

We consider an isothermal adsorption column, packed with porous spherical particles, which is subjected, at time zero, to a small step change in the concentration of an adsorbable species in the carrier. The flow pattern is described by the axial dispersed plug-flow model, and both external and internal diffusional resistances to mass transfer are included so that the system may be described by the following set of equations:

Particle Diffusion:

$$\frac{\partial q}{\partial t} = D\left(\frac{\partial^2 q}{\partial r^2} + \frac{2}{r}\frac{\partial q}{\partial r}\right) \qquad (1)$$

Boundary Conditions:

$$q(r,t = 0) = 0 \qquad (2)$$

$$\left.\frac{\partial q}{\partial r}\right|_{r=0} = 0 \qquad (3)$$

$$D\left.\frac{\partial q}{\partial r}\right|_{r=R} = k\left[c(z,t) - \frac{q|_{r=R}}{K}\right] \qquad (4)$$

External Fluid:

$$-D_L \cdot \frac{\partial^2 c}{\partial z^2} + v\frac{\partial c}{\partial z} + \frac{\partial c}{\partial t} = -\left(\frac{1-\epsilon}{\epsilon}\right)\left\{\frac{3k}{R}\left(c(z,t) - \frac{q|_{r=R}}{K}\right)\right\} \qquad (5)$$

Boundary Conditions:

$$D_L\left.\frac{\partial c}{\partial z}\right|_{z=0} = -v(c|_{z=0} - c|_{z=0+}) \qquad (6)$$

$$\left.\frac{\partial c}{\partial z}\right|_{z=L} = 0 \qquad (7)$$

The problem as defined in Eqs. 1–7 is identical to the problem solved analytically by Rasmuson and Neretnieks (1980) except for the column boundary conditions (Eqs. 6 and 7). These are the correct boundary conditions for a dispersed plug-flow system as discussed by Wehner and Wilhelm (1956). Rasmuson and Neretnieks employed the simpler boundary conditions $c(z = 0,t) = c_o$, $c(z \rightarrow \infty,t) = 0$ used earlier by Rosen (1952). This difference in boundary conditions leads to a significant difference in the solutions only when the axial Peclet number and column length are both small.

Written in dimensionless form Eqs. 1–7 become:

$$\frac{\partial Q}{\partial \tau} = \nabla^2 Q \qquad (8)$$

$$Q(\eta,\tau = 0) = 0 \qquad (9)$$

$$\left.\frac{\partial Q}{\partial \eta}\right|_{\eta=0} = 0 \qquad (10)$$

$$\frac{1}{K}\left.\frac{\partial Q}{\partial \eta}\right|_{\eta=1} = \xi\left\{U - \frac{Q|_{\eta=1}}{K}\right\} \qquad (11)$$

$$\frac{\partial U}{\partial \tau} = \frac{1}{Pe}\psi\theta\frac{\partial^2 U}{\partial x^2} - \psi\theta\frac{\partial U}{\partial x} - 3\psi\xi\left\{U - \frac{Q|_{\eta=1}}{K}\right\} \qquad (12)$$

$$\left.\frac{\partial U}{\partial x}\right|_{x=0} = -Pe\{U|_{x=0-} - U|_{x=0+}\} \qquad (13)$$

$$\left.\frac{\partial U}{\partial x}\right|_{x=1} = 0 \qquad (14)$$