Mass Transfer Studies of External-Loop Airlifts and a Bubble Column

Gas holdups (ϵ_{GR}) and liquid-phase volumetric oxygen transfer coefficients ($k_L a_T$) were measured for a bubble column (BC) and three different external-circulation-loop airlift (ECL-AL) fermentors of 50 to 60 L working volume, using viscous non-Newtonian aqueous solutions of various carboxymethylcelluloses. Some measurements also were done with a viscous Newtonian system (51.8 wt. % sucrose solution).

Discussed in this paper are correlations of ϵ_{GR} and $k_L a_T$ with riser superficial gas velocity ($0.02 \leq u_{GR} \leq 0.26$ m/s), the ratio of the downcomer and riser crosssectional areas (0 [BC] $\leq A_d/A_r \leq 0.444$), and the effective viscosity of the liquid phase ($0.02 \leq \eta_{eff} \leq 0.5$ Pa · s), over the parameter ranges indicated. It is shown that both ϵ_{GR} and $k_L a_T$ are highly dependent upon u_{GR} and A_d/A_r. The effective viscosity has a significant effect on $k_L a_T$, but has only a relatively weak affect on ϵ_{GR}. The $k_L a_T$ correlation developed for non-Newtonian systems was extended to include the results obtained for the viscous Newtonian system studied by incorporating the effects of liquid-phase molecular diffusivity, density, and interfacial tension as determined by Nakanoh and Yoshida (1980).

Milan K. Popovic
Campbell W. Robinson
Department of Chemical Engineering
University of Waterloo
Waterloo, Ontario, Canada N2L 3G1

Introduction

Pneumatically-agitated fermentors increasingly are being considered for use in the fermentation industries in place of the traditional mechanically-agitated bioreactor design because of the potential savings in both capital and operating costs (e.g., power requirement for aeration and mixing). Furthermore, pneumatically-agitated bioreactors are suitable particularly for use with some of the "new" biotechnologies, such as plant cell cultures which employ shear-sensitive cells, because they can exhibit lower shear rates than stirred tanks.

Pneumatic contactors may be classified into two general categories. The first is exemplified by the "bubble-column" (gas-sparged cylindrical vessel of large "aspect ratio," i.e., large height-to-diameter ratio). The second types, known as "airlift" fermentors, are modified bubble columns which are divided (either internally or externally) into two geometrically and hydrodynamically distinct volumetric zones. Under aerated conditions, each zone has a different fractional gas holdup and, hence, a different dispersion density. These two zones are interconnected (either internally or externally); the difference in hydrostatic pressure arising from the difference in dispersion density between the two regions induces liquid circulation in the contactor, thus enhancing the macroscale mixing of the liquid phase compared to that of bubble columns. The zone into which fresh gas is sparged and which has the higher fractional gas holdup is called the "riser" (gas and liquid flow upwards cocurrently), whereas the zone of lower gas holdup (higher dispersion density) is called the "downcomer," wherein gas and liquid flow concurrently downwards.

When the downcomer is located externally to the riser and connected to it by horizontal piping at the top and bottom of the riser dispersion zone, this type is known as the "external-circulation-loop airlift" (ECL-AL), Figure 1. Gas is sparged to the bottom of the riser section (normally the larger diameter column); most of the gas disengages at the top of the riser, but some gas bubbles may be entrained with the liquid flowing through the top horizontal connection into the downcomer section. Liquid (plus entrained gas bubbles) recirculates to the bottom of the riser from the downcomer through the bottom horizontal interconnecting pipe.

For application to aerobic, nonviscous, Newtonian fermentation systems, some previous work has been done on evaluating the gas-liquid oxygen transfer characteristics of ECL-AL fermentors (e.g., Chakravarty et al., 1973; Onken and Weiland,

Present address of M. K. Popovic: Fachhochschule Giessen-Friedberg, Fachbereich Technisches Gesundheitswesen, D-6300 Giessen, Wiesen St. 14, West Germany.
Correspondence concerning this paper should be addressed to C. W. Robinson.

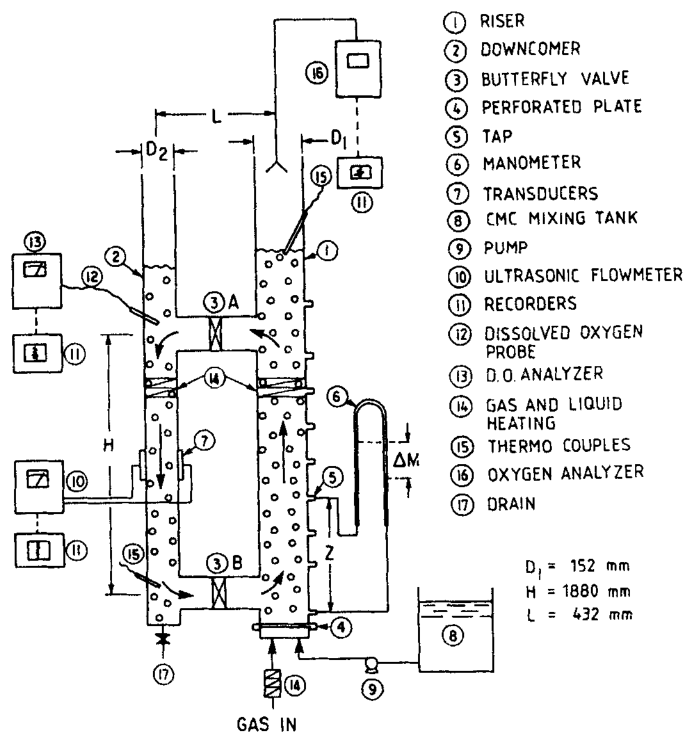

Figure 1. Experimental setup.

1983; Bello et al., 1985a,b; Popovic and Robinson, 1987a). A very limited amount of information is available on their performance with viscous, Newtonian liquids (Onken and Weiland, 1983). Although ECL-AL fermentors have been used for the cultivation of mycelial microorganisms (e.g., Kristiansen, 1978), prior to our recent studies (this work; also Popovic and Robinson, 1984, 1987b) there have been no reported systemic investigations of the mass transfer characteristics of these types of bioreactors with viscous, non-Newtonian systems.

In this present study we have made an extensive investigation of the fractional gas holdup in the riser section (ϵ_{GR}) of various size ECL-AL's, and of the volumetric liquid-phase mass transfer coefficient (k_La_T), using liquid phases having non-Newtonian (power law) viscosity characteristics. For experimental simplicity, we did not use actual mycelial fermentation broths, but instead aqueous solutions of various types of carboxymethylcellulose (CMC), with and without added salt (0.8 kmol/m^3 Na_2SO_3). These systems are chemically similar to those used by us in earlier studies for the determination of the specific interfacial area by the chemical reaction method (Popovic and Robinson, 1987a,b). However, the shear-rate-dependent viscosity range (0.02 – 0.50 Pa · s) studied in this work encompassed the range which is characteristic of actual mycelial fermentations of industrial interest (Stein and Schäfer, 1983; Allen et al., 1987). In addition to varying the effective viscosity and the gas superficial velocity (riser section), for the ECL-AL cases we also studied the effect of varying the geometric configuration as characterized by the ratio of the downcomer and the riser cross-sectional areas (A_d/A_r) in the range $0 \leq A_d/A_r \leq 0.444$ using one riser (0.15 m i.d.) and three different downcomers (0.05, 0.075 and 0.1 m i.d.).

In order to compare our results to those obtained for non-Newtonian liquids (mainly CMC solutions) by previous investigators of bubble column contactors, we also studied the BC mode of operation ($A_d/A_r = 0$). This enabled us to develop correlations of ϵ_{GR} and k_La_T, as discussed later, which are valid for both the BC and the ECL-AL types of pneumatically-agitated fermentors which we studied.

Previous Work

No previous investigators have studied the behaviour of various mass transfer parameters (e.g., ϵ_G, k_La, d_B, etc.) in viscous, non-Newtonian systems in ECL-AL contactors, nor are there existing correlations useful for design and scale-up calculations in such cases. However, numerous studies have been conducted of such parameters and systems in bubble columns (e.g., Buchholz et al., 1978; Voigt et al., 1980; Nakanoh and Yoshida, 1980; Deckwer et al., 1982; Schumpe, 1981; Schumpe and Deckwer, 1982, 1987; Godbole et al., 1982, 1984). Furthermore, some investigations have been made of nonviscous (e.g., Chakravarty et al., 1973; Lin et al., 1976; Onken and Weiland, 1983; Bello et al., 1985a,b) or viscous (Onken and Weiland, 1983) Newtonian systems in loop bioreactors, mainly of the ECL-AL type.

The results of these previous studies suggest that the riser gas holdup (ϵ_{GR}) and the liquid-phase volumetric mass transfer coefficient (k_La_T) are related to the physical properties (ρ_L, σ_L, η_{eff}) and the principal design (A_d/A_r; d_{hole}; H_D) and operating (u_{GR})

parameters by correlations of the types:

$$\epsilon_{GR} = A_1 \rho_L^{a_1} \sigma_L^{b_1} \eta_{\text{eff}}^{d_1} d_{\text{hole}}^{e_1} H_D^{f_1} [(1 + (A_d/A_r)]^{g_1} u_{GR}^{h_1} = G_1 \quad (1)$$

$$k_L a_T = A_2 \rho_L^{a_2} \sigma_L^{b_2} D_L^{c_2} \eta_{\text{eff}}^{d_2} d_{\text{hole}}^{e_2} H_D^{f_2} [(1 + A_d/A_r)]^{g_2} u_{GR}^{h_2} = G_2 \quad (2)$$

where for the bubble column mode of operation $g_1 = g_2 = 0$, since $A_d/A_r = 0$. In this case $u_G = u_{GR}$, $\epsilon_G = \epsilon_{GR}$, and $k_L a = k_L a_T$.

In most previous studies of viscous, non-Newtonian systems, the investigators used a single liquid at only one specified temperature and, thus, the effects of the physical properties (ρ_L, σ_L, D_L) were not determined explicitly. However, Nakanon and Yoshida (1980) found that the correlation for $\epsilon_G[\epsilon_G/(1 - \epsilon_G)^4]$, previously developed for low viscosity Newtonian systems (Akita and Yoshida, 1974), fitted their CMC gas holdups within 30%. The type of CMC used and the consistency and flow behavior indices were not reported for their work. The exponents a_1 and b_1 in Eq. 1 were found to be 0.29 and -0.125, respectively. Nakanoh and Yoshida (1980) also proposed a correlation for $k_L a$ which included the effects of the parameters ρ_L, σ_L and D_L. The exponents a_2, b_2 and c_2 in Eq. 2 were found to be 1.03, -0.75 and 0.5, respectively. They employed a bubble column of 0.1455 m inside diameter and a clear liquid height of 1.1 m, and used single-hole orifice (4 mm dia.) as the gas sparger. The effective viscosity was found to weakly affect both the gas holdup and $k_L a$. The exponents d_1 and d_2 in Eqs. 1 and 2 were reported as -0.17 and -0.28, respectively.

In CMC (Serva, FRG) solutions of different concentrations ($0.04 \leq K \leq 0.23$ Pa · s^n; $0.91 \geq n \geq 0.82$), Deckwer et al. (1982) found no influence of the effective viscosity on bubble column gas holdup, but that it had very strong effect on $k_L a$ [$\propto (\eta_{\text{eff}})^{-0.84}$]. Schumpe and Deckwer (1982) pointed out that the viscosity affects gas holdup only in the churn turbulent flow regime. Both studies were carried out in a 2.7 m high bubble column having an inside diameter of 14 cm and which was equipped with a perforated plate sparger (73 holes of 1 mm diameter). Godbole et al. (1982) measured gas holdup in a 0.305 m diameter bubble column using CMC solution (7H4/Hercules; $0.0018 \leq K \leq 2.57$ Pa · s^n; $1 \geq n \geq 0.495$) and correlated it to $(\eta_{\text{eff}})^{-0.146}$. Repeated measurements two years later (Godbole et al., 1984; CMC types 7M and 7H4/Hercules; $0.095 \leq K \leq 7.68$ Pa · s^n; $0.697 \geq n \geq 0.44$) showed that ϵ_G could be correlated to $(\eta_{\text{eff}})^{-0.19}$ and $k_L a$ to $(\eta_{\text{eff}})^{-1.01}$. Using ϵ_G data obtained in their own work as well as previous works by others, Schumpe and Deckwer (1987) recently presented a gas holdup correlation for the heterogeneous dispersion regime in which $\epsilon_G \propto \rho_L^{0.09}\, \sigma^{-0.13}\, (\eta_{\text{eff}})^{-0.22}\, u_G^{0.54}$. Their correlation is restricted to the cases of $d_{\text{hole}} \geq 1$ mm, $\eta_{\text{eff}} \geq 4$ mPa · s, $u_G \geq 0.02$ m/s, and $u_L \ll u_G$. For the prediction of shear-rate-dependent values of η_{eff}, all the above studies except that of Schumpe and Deckwer (1987) used the shear rate correlation of Nishikawa et al. (1977), which was developed from heat transfer studies of non-Newtonian liquids in a bubble column, i.e.,

$$\dot{\gamma} = \beta \cdot u_G = 5{,}000\, u_G \quad (3)$$

The influence of gas superficial velocity u_{GR} on ϵ_{GR} and $k_L a_T$ depends, at otherwise constant conditions, on the dispersion flow pattern. The effect of u_{GR} in churn-turbulent flow is less pronounced than in the case of either bubbly or slug flow. For churn-turbulent dispersions, Godbole et al. (1984) correlated ϵ_G to $u_G^{0.6}$ and $k_L a$ to $u_G^{0.44}$. For slug flow, Deckwer et al. (1982) correlated ϵ_G to $u_G^{0.82}$ and $k_L a$ to $u_G^{0.59}$. On the other hand, Nakanoh and Yoshida (1980) found $k_L a'$ to be proportional to u_G, which observed behaviour differs significantly from the results of all other authors.

Schumpe and Deckwer (1987) recently reviewed the hydrodynamic and mass transfer characteristics of viscous media in bubble columns. In addition, they presented new experimental data for viscous liquid systems (aqueous solutions of glycerol, CMC, polyacrylamide and xanthan gum, the latter three types of solutions being non-Newtonian). Two different bubble columns were used, each being equipped with a ring sparger: one of $D = 14$ cm and $H_D = 2.2$ m (sparger having 29 2 mm dia. holes), the other of $D = 30$ cm and $H_D = 2.0$ m (sparger having 56 2 mm dia. holes). The new mass transfer data, plus previous data obtained by Schumpe and Deckwer (1982) and by Deckwer et al. (1982) was used to develop the following correlation for $k_L a$, based on the functional relationships previously obtained by Nakanoh and Yoshida (1980) from dimensional analysis:

$$k_L a D^2/D_L = 0.021(\eta_{\text{eff}}/\rho_L D_L)^{0.5}(gD^2\rho_L/\sigma)^{0.21} \cdot (gD^3\rho_L^2/\eta_{\text{eff}}^2)^{0.60}[u_G/(gD)^{0.5}]^{0.49} \quad (4)$$

Equation 4 described the experimental data with a mean deviation of 15.8%. The best fit of the data was obtained using effective viscosities for the pseudoplastic fluids based on $\beta = 2{,}800$ m^{-1} in Eq. 3 for the prediction of shear rate. From Eq. 4, it follows that

$$k_L a \propto u_G^{0.49} D^{-0.025} D_L^{0.50} (\eta_{\text{eff}})^{-0.70} \rho_L^{0.91} \sigma^{-0.21} \quad (5)$$

Equations 4 and 5 are said to be applicable to both the heterogeneous and the slug-flow regimes (Schumpe and Deckwer, 1987).

To date, the effect of sparger type (d_{hole} in Eqs. 1 and 2) has not been clearly determined quantitatively. Directionally, both ϵ_G and $k_L a$ increase with decreasing d_{hole} in the bubbly flow regime. In most cases, H_D also was not varied such that its effect, similar to that of constant physical properties (ρ_L, σ_L, D_L), merely was incorporated in the values of A_1 and/or A_2 in Eqs. 1 and 2, respectively. However, Bello (1981) showed theoretically and provisionally verified by comparison of his results ($H_D = 1.8$ m) to those of Onken and Weiland (1980), who used an ECL-AL having $H_D = 8.5$ m, that the circulating liquid velocity (u_{LR}) in an ECL-AL fermentor increases with increasing H_D at constant A_d/A_r according to $u_{LR} \propto (H_D)^{0.333}$. It is known for Newtonian systems that u_{LR} increases with increasing A_d/A_r at otherwise constant conditions (Bello et al., 1984) and that both ϵ_{GR} and $k_L a_T$ decrease with increasing u_{LR} (Onken and Weiland, 1980; Bello et al., 1984). Thus, the results of experiments at constant H_D produce correlations of the types of Eqs. 1 and 2 where in effect $f_1 = f_2 = 0$, but in which H_D may have influenced the determined values of exponents g_1 and g_2 for the ECL-AL cases.

Bello (1981) correlated gas holdup data from water and water/salt solution to $(u_{GR}/u_{LR})^{0.56}(1 + A_d/A_r)$ and $k_L a$ data to $u_{GR}^{0.8}\,(1 + A_d/A_r)^{-2}$. Onken and Weiland (1983) carried out six measurements in 34.7 wt. % saccharose ($\mu = 4$ mPa · s) and four measurements in 51.8 wt. % saccharose solution ($\mu = 16$ m · Pa · s). They found a pronounced effect of viscosity on $k_L a$ in these viscous Newtonian systems. The values of the coefficients

Table 1. Values of the Coefficients and Exponents in Eqs. 1 and 2 for Previous Correlations

Reference	A_1	a_1	b_1	d_1	e_1	f_1	g_1	h_1
Akita and Yoshida (1973)*	0.2	0.29	−0.125	−0.17	—	—	—	1
Bello et al. (1985a)**	0.16	—	—	—	—	—	1	0.56
Deckwer et al. (1982)	1.16	—	—	—	—	—	—	0.82
Godbole et al. (1982)	0.255	—	—	−0.146	—	—	—	0.532
Godbole et al. (1984)	0.255	—	—	−0.19	—	—	—	0.6
Nakanoh and Yoshida (1980)†	—	—	—	—	—	—	—	—
Schumpe (1981) (Schumpe and Deckwer, 1982 for $D = 0.14$ m)	0.718	—	—	—	—	—	—	0.674
Schumpe and Deckwer (1987)‡	0.20	0.09	−0.13	−0.22	—	—	—	0.54

Reference	$A_2 \times 10^3$	a_2	b_2	c_2	d_2	e_2	f_2	g_2	h_2
Akita and Yoshida (1973)*	—	—	—	—	—	—	—	—	—
Bello et al. (1985a)**	7.60	—	—	—	—	—	—	−2	0.8
Deckwer et al. (1982)	3.15	—	—	—	−0.84	—	—	—	0.59
Godbole et al. (1982)	—	—	—	—	—	—	—	—	—
Godbole et al. (1984)	0.835	—	—	—	−1.01	—	—	—	0.44
Nakanoh and Yoshida (1980)†	90	1.03	−0.75	0.5	0.28	—	—	—	1
Schumpe (1981) (Schumpe and Deckwer, 1982 for $D = 0.14$ m)	—	—	—	—	—	—	—	—	—
Schumpe and Deckwer (1987)‡	21	0.91	−0.21	0.50	−0.70	—	—	—	0.49

*$\epsilon_G/(1-\epsilon_G)4_g 0.29 = G_1$
**$\epsilon_G \cdot u_{LR} 0.56 = G_1$
†$k_L a'/D^{0.17} g^{0.64} = G_2$
‡G_1 for heterogeneous regime; $\epsilon_G g^{0.29} D^{0.20} = G_1$

and exponents in Eqs. 1 and 2 for the previous correlations are summarized in Table 1.

Materials and Methods

Apparatus

The external-circulation-loop airlift bioreactor used in this work is illustrated schematically in Figure 1. It consisted of the riser (i.d. 0.15 m), item 1 in the figure, and downcomer (item 2), both made of Pyrex glass. Three different sized downcomers, having inside diameters of 0.05 m, 0.075 m, and 0.10 m were employed during the measurements. Only one of these downcomers is shown on Figure 1 for clarity. In this way, the values of the downcomer-to-riser cross-sectional area ratios (A_d/A_r) which were investigated were 0.111, 0.25, and 0.44. Each downcomer was linked to the riser by means of a pair of butterfly valves (items 3A and 3B). Closing both valves converted the ECL-AL to a simple bubble column (BC; $A_d/A_r = 0$). By means of throttling the top or bottom valve the liquid circulation velocity could be varied or interrupted.

A perforated stainless steel plate, with 52 1 mm dia. holes (triangular pitch), was used as the sparger. The gas supply network (not depicted) consisted of two lines (for air and N_2), two needle valves and a calibrated rotameter. The sparged gas was preheated (item 14 in Figure 1) to the column operating temperature ($T = 23$°C), which was maintained constant by means of heating tapes installed on the riser and the downcomer (item 14 in Figure 1). The test liquids were prepared in a tank (item 8) equipped with a stirrer and cooling or heating coils (not depicted), and were transported to the contactor by a pump (item 9). All experiments were conducted in the semibatch mode (batch liquid; continuous gas sparging) with a liquid volume of 50 to 60 L. All runs were made at local barometric pressure (riser gas headspace).

The circulating liquid superficial velocity in the riser (u_{LR}) was determined by measuring the corresponding value in the downcomer (u_{LD}) and using the liquid-phase continuity equation

$$u_{LR} = u_{LD}(A_d/A_r) \tag{6}$$

An ultrasonic flow-meter which responds to the downflowing small-bubble fraction in the central core of the downcomer was used to measure u_{LD} (Popovic and Robinson, 1988).

Gas holdups (riser and downcomer) measurements

Gas holdups were measured using the volume expansion method. After reading the dispersion height (H_D) in the ECL-AL riser and downcomer during aeration, valves (item 3 in Figure 1), were closed rapidly and simultaneously and the gas supply was stopped (for the BC case, valves 3A and 3B already were tightly shut). Ten minutes later, the gas entrapped around valves 3A and 3B was released by means of small vents (not depicted in Figure 1); the clear liquid height (H_L) in both parts of the ECL-AL fermentor or in the BC then was measured and corrected for the fraction of small entrapped bubbles still retained by the liquid. This "small bubble volume fraction" was measured by manometers attached to the riser and the downcomer (item 6 in Figure 1). Gas holdups then were computed from

$$\epsilon_{Gi} = (H_{Di} - H_{Li})/H_{Di} \qquad i = R, D \tag{7}$$

For an airlift fermentor the total gas holdup is equal to the sum of the gas volumes in the two principal sections:

$$\epsilon_{GT} = \epsilon_{GR}(V_{DR}/V_{DT}) + \epsilon_{GD}(V_{DD}/V_{DT}) \tag{8}$$

Volumetric oxygen transfer coefficient (k_L/a_T) measurement

Volumetric mass transfer coefficients were determined using the dynamic method described by Nakanoh and Yoshida

(1980). For this purpose, the liquid was deaerated by sparging nitrogen. After stopping the nitrogen flow and after allowing disengagement of the nitrogen bubbles, the air flow was started, and the increase in liquid-phase dissolved oxygen tension was measured with a dissolved oxygen electrode (Yellow Springs Inst. Co., OH; Model YSI 5739, item 12 in Figure 1) connected to a dissolved-oxygen analyzer (YSI Model 57-YSI, item 13 in Figure 1). The fast-response D.O. probe was located in either the upper cross piece region of the riser (in which case it was positioned with the membrane pointing up to avoid bubbles collecting on the membrane surface), or in the upper tee piece region of the downcomer. When used with very viscous liquids, a tiny stirrer was mounted adjacent to the D.O. probe membrane in order to minimize the effects of the liquid-phase hydrodynamic boundary layer mass transfer resistance on the D.O. probe response.

Assuming a well-mixed liquid phase and a negligible change in gas concentration along the column height, the unsteady state liquid-phase mass balance for oxygen can be written as:

$$V_L(dC_L/dT) = k_L F_T(C^* - C_L) \tag{9}$$

where F_T is the total specific interfacial area effective for mass transfer. In an ECL-AL reactor, the total liquid volume consists of the sum of that in the riser and the downcomer. Therefore,

$$V_L = V_{LR} + V_{LD} \approx H_L(A_r + A_d) \approx H_L A_r[1 + (A_d/A_r)] \tag{10}$$

In the riser section there is intensive slip between the phases compared to the essentially no-slip situation in most of the downcomer (Popovic and Robinson, 1988). Therefore, it is realistic to assume that the major part of the mass transfer-active gas-liquid interfacial area is located in the riser, and thus

$$F_T \approx F_R \tag{11}$$

Hence Eq. 9 becomes

$$\begin{aligned}[1 + (A_d/A_r)](dC_L/dt) &= k_L(F_R/H_L A_r)(C^* - C_L) \\ &= k_L a'_R(C^* - C_L)\end{aligned} \tag{12}$$

Integration of Eq. 12 under the assumption C^* is constant along the riser axis, and using the initial condition $C_L = C_{L0}$ at $t = 0$, results in

$$\ln[(C^* - C_L)/(C^* - C_{L0})] = k_L a'_T t \tag{13}$$

where $k_L a'_T$ is defined as

$$k_L a'_T = k_L a'_R[1 + (A_d/A_r)]^{-1} \tag{14}$$

Plotting $\ln(C^* - C_L)$ against time (t) gives as the slope the overall mass transfer coefficient ($k_L a'_T$). From the $k_L a'_T$ values the volumetric mass transfer coefficients with respect to dispersion volume ($k_L a_T$) were calculated as:

$$k_L a_T = k_L a'_T(1 - \epsilon_{GT}) \tag{15}$$

In the BC mode of operation, $A_d = 0$ and

$$k_L a'_T = k_L a'_R = k_L a' \tag{16}$$

and

$$\epsilon_{GT} = \epsilon_{GR} = \epsilon_G \tag{17}$$

The assumption about an essentially well mixed liquid for the purpose of $k_L a$ measurement also was made by Nakanoh and Yoshida (1980), and was verified by Patwari (1983). Patwari used the bubble column and the same type of CMC solutions as described by Deckwer et al. (1982). Patwari (1983) estimated $k_L a$ values by fitting an axial dispersion model to measured steady state oxygen profiles and compared these results to those obtained using the dynamic method as used here; no differences were observed. The liquid mixing in an ECL-AL fermentor is even better than in a bubble column of the same aspect ratio (Onken and Weiland, 1980; Popovic and Robinson, 1988). The assumption of negligible change in the gas concentration was proved by measuring the oxygen concentrations of the inlet and outlet air-streams, using a paramagnetic oxygen analyzer (Beckman Instruments, Fullerton, CA, Model 755A; item 16 in Figure 1). The validity of Eqs. 11 and 14 will be discussed later.

Systems studied

Dilute solutions of CMC frequently have been used as model media for filamentous microbiological cultures, but significantly different results for ϵ_G and $k_L a$, measured using CMC solutions in geometrically-similar bubble columns and under the same operating conditions, have been reported (Deckwer et al., 1982; Buchholz et al., 1978; Nakanoh and Yoshida, 1980). One of the possible sources of these discrepancies may be differences in the physicochemical properties (rheological behavior, surface tension, etc.) of the various kinds of CMC previously used. Therefore, several types of CMC were used in this present study in an attempt to understand some of the previous discrepancies. The CMC types used by us and their rheological properties (for 1.0% w/w solutions as an example) are listed in Table 2. It should be noted that most of the present work was done using solutions of different concentrations of Hercules 7H3XF CMC.

The rheological behaviour of the CMC solutions was measured using a rotoviscometer (Fann, Model 35A). All solutions were found to obey the power law relationship between shear rate ($\dot{\gamma}$) and shear stress (τ), so that the effective viscosity could be calculated as follows:

$$\eta_{\text{eff}} = \tau/\dot{\gamma} = K(\dot{\gamma})^{n-1} \tag{18}$$

Values of the rheological constants (K, n) are given in the various figures. The correlation of Nishikawa et al. (1977), i.e., Eq. 3, was used to estimate the shear rate ($\dot{\gamma}$). Equation 3 was found by Nishikawa et al. (1977) to be independent of liquid superficial velocity for $u_L \le 0.14$ m/s, the maximum u_L which they tested. In this present work, we observed values of u_{LR} of up to 0.35 m/s, but assumed that Eq. 3 remained valid in the range $0.14 \le u_{LR} \le 0.35$ m/s. The viscosity range covered in this work was $0.02 \le \eta_{\text{eff}} \le 0.5$ Pa · s, being achieved over the gas superficial velocity range $0.02 \le u_{GR} \le 0.26$ m/s for both the ECL-AL and BC modes of operation. Surface tensions (Table 2) were measured using a ring tensiometer (Fischer Tensiomat 21), and the solution densities using a specific gravity bottle.

In addition to the viscous, non-Newtonian liquids tested, for comparative purposes we also made some measurements using a

Table 2. Physical Properties at 23°C of Aqueous CMC, CMC/Salt and Sucrose Solutions Used in This Work

Type/ Company	Conc. (wt. %)	Consistency Index (Pa · s^n)	Flow Behavior Index	Surface Tension (N/m) × 10^3	Liquid Phase Diffusivity* (m^2/s) × 10^9	Density (kg/m^3)	CMC Type Previously Used by:
7H3SXF/Hercules	1	2.03	0.57	75.5	2.30	1,004	Popovic & Robinson (1987b, 1988)
7H4/Hercules	1	9.2	0.4	79.3	2.30	1,009	Godbole et al. (1982)
7M/Hercules	1	0.19	0.69	75.4	2.30	1,002	Godbole et al. (1984)
Tylose C300/Hoechst	1	0.18	0.78	62.1	2.30	1,007	Buchholz et al. (1978)
Serva/Heidelberg	1	0.15	0.79	71.4	2.30	1,004	Schumpe & Deckwer (1982)
CRT 5000/Bayer	1	0.99	0.63	70	2.30	1,003	Onken & Weiland (1980)
Relatin U3000/Henkel	1	0.069	0.71	61.5	2.30	1,012	Deckwer et al. (1982)
7H3SXF/Hercules in 0.8 kmol/m^3 Na_2SO_3 Solution	0.5	0.25	0.63				Popovic & Robinson (1987b)
	1.0	0.90	0.66				
	1.5	3.5	0.61				
51.8 wt. % Sucrose Solution		μ = 0.019 Pa · s		74.56	0.31	1,240.6	Onken & Weiland (1980)

*From Navari et al. (1971) and Hikita et al. (1978)

viscous, Newtonian liquid (51.8 wt. % sucrose solution). Its viscosity, interfacial tension and density were determined as described above for the CMC solutions.

Results and Discussion

Two-phase flow patterns

Because of the wide range of operating conditions, e.g., $0.2 \le u_{GR}$[m/s] ≤ 0.26, and the different CMC solutions ($0.05 \le K$[Pa · s^n] ≤ 9.3) used in the present study, it is difficult to unambiguously define the flow pattern in the riser (upflow) as being the same in all cases. For the liquids of smaller effective viscosity (i.e., below about 0.04 Pa · s), large irregular-shaped bubbles characteristic of the churn-turbulent flow regime were observed. At higher effective viscosities and/or higher gas velocities, cap bubbles indicative of the transition to the slug flow regime prevailed. Finally, for the highest viscosity liquids ($\eta_{eff} \ge$ 0.1 Pa · s), bullet-shaped wall-to-wall bubbles characteristic of the fully developed slug flow regime according to Devine et al. (1985) were observed at all gas velocities. Seventy-five percent of all runs in the present work were carried out in fully developed slug flow ($\eta_{eff} \ge 0.1$ Pa · s) and 90% at $\eta_{eff} > 0.04$ Pa · s.

In contrast to the flow patterns observed in the riser, those in the downcomer were uniform over the entire range of η_{eff} and u_{GR} examined. The larger bubbles carried over from the riser escaped immediately at the top of downcomer. The smaller entrained bubbles at first migrated to the wall of the downcomer and then moved back to assume the coring bubble pattern in downward flow. At high gas throughputs, bubble clusters coming from the bottom horizontal piping occasionally were observed to creep upwards along the downcomer wall. Over the entire operating range studied, the liquid flow, especially in the bottom half of downcomer, was straight down, and was free of any turbulence. The downcomer liquid Reynolds number (Re_L) was always less than 2,300.

Liquid velocity in the riser (ECL-AL)

As we recently have shown elsewhere (Popovic and Robinson, 1988), the velocity of the circulating liquid in the riser increases with increasing u_{GR} and A_d/A_r and with decreasing η_{eff} according to

$$u_{LR} = 0.23\, u_{GR}^{0.32} (A_d/A_r)^{0.97} (\eta_{eff})^{-0.39} \tag{19}$$

Equation 19 is valid when both valves 3A and 3B on Figure 1 are wide open. For the same operating conditions and rheological behaviour (e.g., CMC having $K = 1.6$ Pa · s^n and $n = 0.59$), u_{LR} with $A_d/A_r = 0.444$ is approximately three times greater than obtained at $A_d/A_r = 0.111$, e.g., 11.5 vs. 3.5 cm/s, respectively, at $u_{GR} = 10$ cm/s. The fact that the upward velocity of the liquid in the riser (cocurrent to the riser gas velocity) increases significantly with the increasing u_{GR} and/or A_d/A_r should be kept in mind when considering the gas holdup and mass transfer coefficient behaviour discussed below.

Riser gas holdup

Part of the riser gas holdup results of our investigations (around 250 measurements were carried out) is presented in Figure 2. For viscous non-Newtonian liquids the results show that ϵ_{GR} is influenced mostly by the superficial gas velocity (u_{GR}). However, the influence of u_{GR} in the heterogeneous flow regime is less pronounced than in bubbly flow. The slope of the ϵ_{GR} vs. u_{GR} line (Figure 2) decreases for $u_{GR} \ge 0.03$ m/s, where

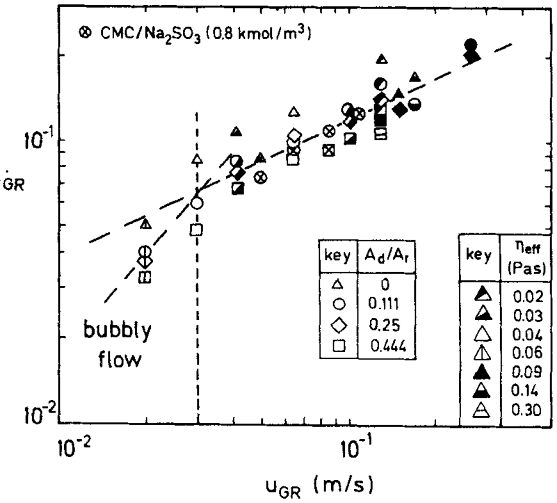

Figure 2. Gas holdup (ϵ_{GR}) in the riser of the ECL-AL fermentor for different A_d/A_r ratios and in the bubble column ($A_d/A_r = 0$) as a function of gas superficial velocity (u_{GR}).

onset of churn-turbulent flow is assumed. While gas holdup increases strongly with increasing u_{GR}, there is only a weak influence of effective viscosity. For example, the 15-fold decrease of η_{eff} at $u_{GR} = 0.134$ m/s (i.e., from 0.3 to 0.02 Pa · s on Figure 2) causes an increase in riser gas holdup of only 20%.

As shown in Figure 2, addition of Na_2SO_3 at a concentration of 0.8 kmol/m^3 gives the same ϵ_{GR} as measured in salt-free CMC solutions at the same operating conditions. Schumpe and Deckwer (1982) also reported that gas holdups, measured under similar operating conditions as in the present work, were the same in CMC and CMC/salt solutions. The effect of reactor geometry also is obvious from Figure 2. As the downcomer diameter increases, that is as A_d/A_r increases (which results in increased u_{LR}), the riser gas holdup decreases. One of the reasons for this influence is the effect of u_{LR} on ϵ_{GR}. Because of the cocurrent gas-liquid flow, the gas bubbles in the riser of the ECL-AL will rise faster, their residence time will decrease and, consequently, riser gas hold-up will decrease. However, plotting the gas holdups measured only in ECL-AL fermentors against the circulating liquid velocity (Figure 3) revealed a more complex nature of the ϵ_{GR} vs. A_d/A_r dependency. From Figure 3 it is obvious that ϵ_{GR} is rather weakly dependent on u_{LR}. An increase in u_{LR} from 0.066 to 0.25 m/s decreases ϵ_{GR} by only 22%. Regressional analyses showed that

$$\epsilon_{GR} \propto u_{LR}^{-0.134} \tag{20}$$

and that the dependency of ϵ_{GR} on A_d/A_r (excluding the BC case, $A_d/A_r = 0$) was

$$\epsilon_{GR} \propto (A_d/A_r)^{-0.129} \tag{21}$$

Using the correlation for circulating liquid velocity (Eq. 19), it is possible to show that Eqs. 20 and 21 are consistent.

The gas holdups measured in CMC/water solutions (using all the types of CMC listed in Table 1) as well as in CMC/salt (0.8 kmol/m^3 Na_2SO_3) and in the sucrose solution were correlated by

$$\epsilon_{GR} = 0.465\, u_{GR}^{0.65}[1 + (A_d/A_r)]^{-1.06}(\eta_{eff})^{-0.103} \tag{22}$$

For the bubble column case, $A_d/A_r = 0$ and $\epsilon_{GR} = \epsilon_G$ in Eq. 22. Equation 22 fits the measured data with a mean deviation of ±11%. The riser gas holdups (measured and predicted) are given in Figure 4 together with the data of Onken and Weiland (1980) for viscous, Newtonian solution. The measured gas holdups of Onken and Weiland (1980) on the average are 27% lower than predicted by Eq. 22. This difference can be attributed to the higher liquid velocity, caused by the greater column height in their case (Onken and Weiland, 1980). However, using the semiempirical relationship developed by Bello (1980), namely $u_{LR} \propto H_{DR}^{0.333}$, in conjunction with Eq. 20 it is not possible to explain the difference observed between the measured (Onken and Weiland, 1980) and predicted (Eq. 22) gas holdups. Therefore, for scale-up purposes, the effect of H_D on ϵ_{GR} remains an unresolved question.

The dependencies of ϵ_{GR} on u_{GR} and η_{eff} shown in Eq. 22 differ somewhat from those given in the recent BC correlation of Schumpe and Deckwer (1987), who found $\epsilon_G \propto u_G^{0.54}\, \eta_{eff}^{-0.22}$. The differences in the values of the two exponents between our correlation (Eq. 22) and theirs may be due to the fact that to calculate η_{eff} we used $\beta = 5{,}000$ m^{-1} in Eq. 3, according to the study of Nishikawa et al. (1977), whereas Schumpe and Deckwer (1987) used a fitted value of $\beta = 2{,}800$ m^{-1}. However, differences in the calculated values of ϵ_G for the BC case using Eq. 22 and the new correlation of Schumpe and Deckwer (1987) are small, since the latter fitted well the data of Schumpe and Deckwer (1982), which in turn agrees quite well with predictions based on Eq. 22, Figure 5.

Measured gas holdups for the bubble column mode of operation from the present work (ϵ_G, meas) and different predictions from the literature are compared in Figure 5. The referred cor-

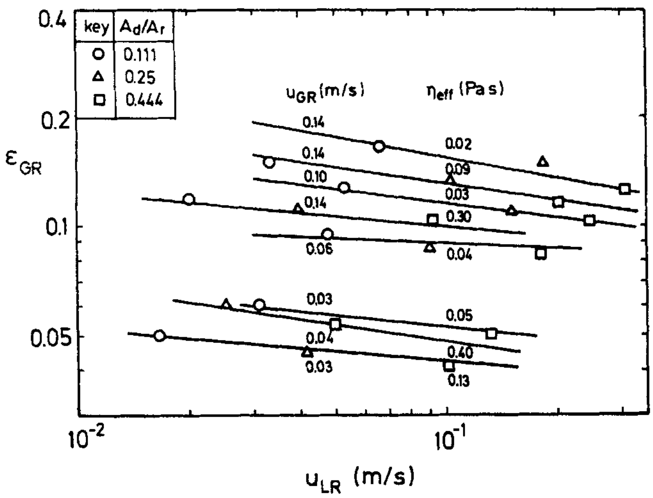

Figure 3. Gas holdup (ϵ_{GR}) as a function of liquid circulating velocity (u_{LR}) in the riser of the ECL-AL fermentor at different A_d/A_r ratios, gas superficial velocities (u_{GR}), and effective viscosities (η_{eff}).

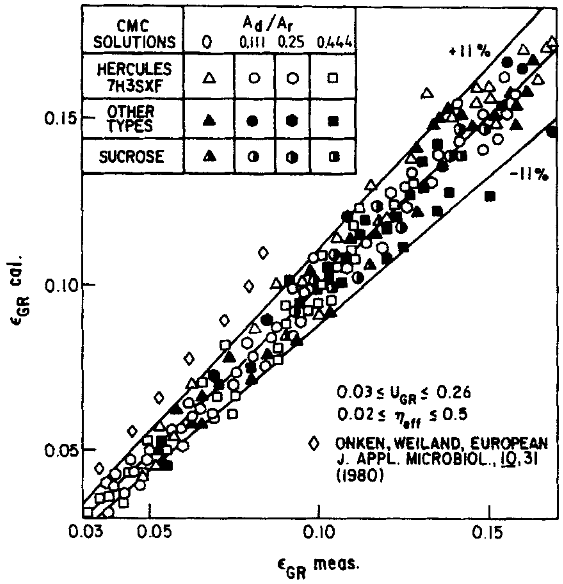

Figure 4. Parity plot of gas holdup correlation, Eq. 22, for viscous liquids in BC and ECL-AL fermentors. Comparison with data of Onken and Weiland (1980; 51.8 wt. % sucrose solution).

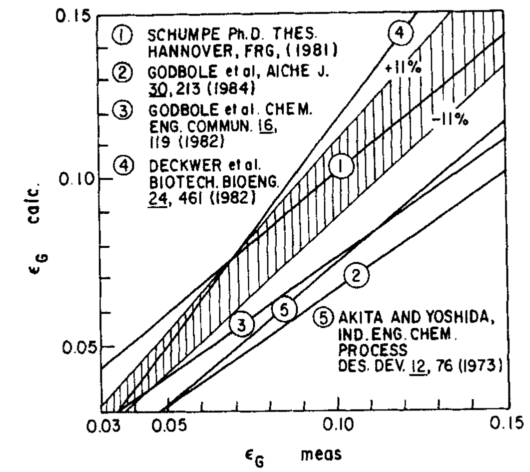

Figure 5. Comparison of the measured gas holdup (bubble column) from present work and the predictions from literature.

relations can be obtained from Table 1, using Eq. 1. The best fit of our measured gas holdups was achieved by the correlation of Schumpe (1981), later published together with Deckwer (Schumpe and Deckwer, 1982). The correlation of Deckwer et al. (1982) predicts slightly higher gas holdups than found in the present work. Both investigations (Schumpe and Deckwer, 1982; Deckwer et al., 1982) were carried out in a 0.14 m diameter bubble column. Godbole et al. (1982, 1984) observed markedly smaller gas holdups in a 0.3 m diameter bubble column. The effect of the gas velocity (u_G) was found to be weaker and that of the effective viscosity more pronounced (see Table 1) than in Eq. 22. The reason for these differences is the different flow pattern which existed in the column of Godbole et al. (1982, 1984). At moderate gas throughouts (comparable to those of the present work), their larger column diameter led to the prevailing flow pattern being churn turbulent in their case, compared to the possible transition to slug flow and partially- or fully-developed slug flow in our work. The correlation of Akita and Yoshida (1973), applied later by Nakanoh and Yoshida (1980), strongly underestimates our measured gas holdups, Figure 5. Considering also the large deviation of the Nakanoh and Yoshida (1980) predictions from those of Schumpe (1981) and Deckwer et al. (1982), as shown on Figure 5, the Akita-Yoshida (1973) correlation cannot be recommended for gas holdup prediction in non-Newtonian dispersions, at least when perforated-plate spargers are employed.

Bello (1981) measured gas holdup in an ECL-AL (geometrically identical to that used in the present work) using water and water/salt solution (0.1 kmol/m^3 NaCl). Employing four different size downcomers ($0.111 \leq A_d/A_r \leq 0.69$) and varying the riser superficial gas velocity (u_{GR}) between 0.014 and 0.086 m/s, he found that the gas holdup in the riser could be predicted by

$$\epsilon_{GR} \propto (u_{GR}/u_{LR})^{0.56}[1 + (A_d/A_r)] \tag{23}$$

The data set of u_{LR} considered by Bello (1981) for his ϵ_{GR} correlation (Eq. 23) was obtained with fully-open valves (such as items 3A and 3B on Figure 1). In this case u_{LR} is a nonadjustable parameter dependent on the A_d/A_r ratio and on u_{GR}. Therefore, u_{LR} is not known *a priori* and is not suitable as a parameter for correlation. However, using Bello's (1981) data for u_{LR} it is possible to convert Eq. 23 to a more practical form. The u_{LR} values obtained by Bello (1981) at $u_{GR} > 0.0346$ m/s follow the dependency

$$u_{LR} \propto [1 + (A_d/A_r)]^{3.68} \tag{24}$$

Substitution of Eq. 24 in Eq. 23 leads to

$$\epsilon_{LR} \propto u_{GR}^{0.56}[1 + (A_d/A_r)]^{-1.06} \tag{25}$$

which is in good agreement with our ϵ_{GR} correlation, Eq. 22, with respect to the effect of u_{GR}, and gives an identical dependency on A_d/A_r.

Downcomer gas holdup

Typical downcomer gas holdup data necessary for the evaluation of the liquid-phase volumetric mass transfer coefficients based on dispersion volume (k_La_T) using Eqs. 8 and 15 are given on Figure 6. For specified geometrical configuration at the top interconnecting section of an ECL-AL, the gas holdup in the downcomer (ϵ_{GD}) depends on the gas holdup and bubble size distribution in the riser, as well as on the liquid velocity in the downcomer (Popovic and Robinson, 1988). All these three parameters are strongly affected by the gas superficial velocity, u_{GR}. This interactive effect of u_{GR} results in having a stronger dependency of the downcomer gas holdup on u_{GR} than that of the riser gas holdup. Similar results were obtained by Chakravarty et al. (1973) in their concentric tube airlift fermentor.

For the given geometrical conditions and physical properties of the CMC solutions used by us, the bubble rise velocity in the downcomer usually was less than 10^{-2} m/s and, hence, was negligibly small in comparison to the downward liquid velocity, u_{LD}, which ranged from 0.08 to 0.5 m/s (Popovic and Robinson, 1988). Therefore, it is reasonable to assume the absence of any slip between the gas phase (entrained bubbles) and the liquid in

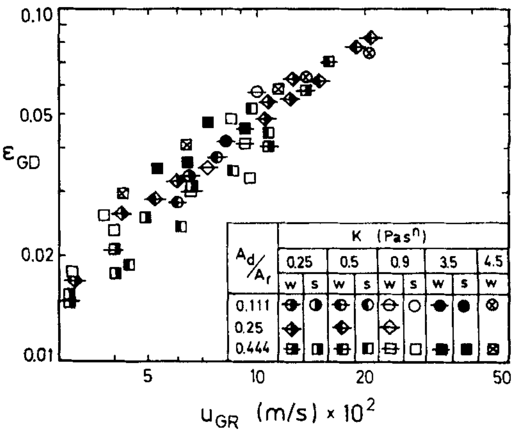

Figure 6. Downcomer gas holdups (ϵ_{GD}) measured in CMC/water solutions (*W*) and in CMC/salt (0.8 kmol/m^3 Na_2SO_3) solutions (*S*) at different A_d/A_r ratios and consistency indices (*K*) as function of the riser gas superficial velocity (u_{GR}).

the downcomer. As a consequence, the interphase oxygen transfer rate in the downcomer is negligible, which confirms the validity of Eq. 11.

Volumetric mass transfer coefficient

As described previously, we measured $k_La'_T$ for oxygen transfer using the dynamic method developed by Yagi and Yoshida (1975) and Nakanoh and Yoshida (1980). Therefore, the first logical step in evaluating our data (particularly for the BC cases) was comparison with the predicted k_La' values of the correlation of Nakanoh and Yoshida (1980; see Table 1). For specified values of the physical properties of CMC solution (D_L, ρ_L, σ_L) their correlation, developed for bubble columns, can be written solely in terms of the superficial gas velocity and the effective viscosity, as shown on Figure 7. For the bubble column case ($A_d/A_r = 0$), predictions of $k_La'_T$ based on the Nakanoh and Yoshida (1980) correlation highly overestimate our experimental results. One of the reasons for this overestimation is the high value of the exponent (1.0) on their u_G term. In addition, the value of their exponent on the effective viscosity term (-0.28) also appears to be incorrect. Curves 1 and 2 on Figure 7 demonstrate this; for $A_d/A_r = 0.111$, increasing the consistency index from $K = 0.21$ to $K = 5$ Pa · s^n(D_L, ρ_L and σ_L remain approximately constant, see Table 2) results in a marked increase of deviation from the predicted values. Although as shown by Eq. 14, at the same u_{GR} and u_{LR} in both cases $k_La'_R$ for an ECL-AL bioreactor ($A_d/A_r > 0$) will always be less than the corresponding $k_La'_R$ in a BC ($A_d/A_r = 0$), due to the "dead" volume with respect to mass transfer in the downcomer of the ECL-AL. Therefore, predictions based on the BC ($u_{LR} = 0 = A_d/A_r$) correlation of Nakanoh and Yoshida (1980) are expected to overpredict $k_La'_T$ values obtained in ECL-AL contactors. However,

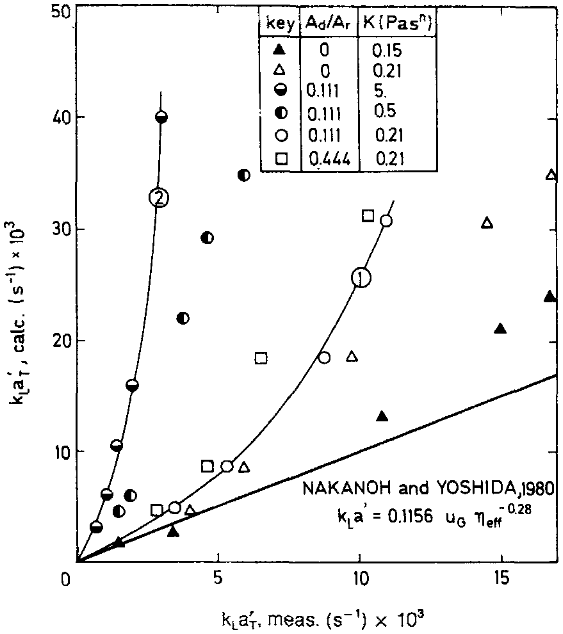

Figure 7. Comparison of the volumetric mass transfer coefficients measured in BC and ECL-AL fermentors (k_La', meas.) with the BC predictions of Nakanoh and Yoshida (1980).

for curves 1 and 2 on Figure 7 $(1 + A_d/A_r) = 1.111$ and with this smallest downcomer u_{LR} is relatively small (on the order of 2 and 7 cm/s for curves 2 and 1, respectively, compared to $u_{LR} = 0$ for the BC case), whereas the predictions deviate up to 3- to 10-fold from the observed $k_La'_T$ values. Therefore, the deviations cannot be attributed solely to geometric differences between the BC and the ECL-AL cases.

As also shown on Figure 7, there is a strong effect of the mode of operation (BC or ECL-AL) on $k_La'_T$; $k_La'_T$ increases with decreasing A_d/A_r, approaching a maximum value for $A_d/A_r = 0$ (BC mode) for given operating conditions (u_{GR}, η_{eff}). This may have been caused by two factors:

1) As the downcomer size increases, the fraction of the total liquid volume which is in the downcomer increases. As discussed previously, the downcomer can be regarded as a "dead" mass transfer zone.

2) The increased liquid velocity with increased A_d/A_r ratio (Eq. 14) acts to lower the riser gas holdup and, hence reduces the specific interfacial area (a') and k_La' in the riser section.

In Table 3 the results of two characteristic runs at high u_{GR} and low η_{eff}, and *vice versa*, demonstrate the validity of the first assumption (i.e., validity of Eqs. 11 and 14); at specified u_{GR}, the observed decrease in k_La_T is directly proportional to the increase in $(1 + A_d/A_r) \approx V_{LT}/V_{LR}$. On the other hand, Table 3 discloses a weak influence of the circulating liquid velocity (u_{LR}) on $k_La'_T$. The approximately fourfold change in u_{LR} shown does not have a significant additional effect on $k_La'_T$. Regression analysis using k_La_T values (k_La_T values are reported more commonly than $k_La'_T$ in the literature) showed that they depended on u_{LR} according to $k_La_T \propto u_{LR}^{-0.07}$. Thus, the conclusion in the review paper by Shah et al. (1982) about the negligible influence of u_L on k_La for low-viscosity Newtonian systems and liquid velocities up to 0.108 m/s, can be extended to viscous non-Newtonian systems at higher liquid velocities (up to 0.34 m/s).

The initial attempt to correlate the measured k_La_T values (using Hercules CMC type 7HSXF) to u_{GR}, $(1 + A_d/A_r)$ and η_{eff} resulted in the following:

$$k_La_T = (2.14)(10^{-3})u_{GR}^{0.52}[1 + (A_d/A_r)]^{-0.85}(\eta_{eff})^{-0.89} \quad (26)$$

Measured k_La_T values in the 51.8 wt. % sucrose solutions followed the dependencies on u_{GR}, $(1 + A_d/A_r)$ and η_{eff} given by Eq. 26, but the absolute k_La_T values were always lower than predicted. That deviation may be attributed to the lower liquid

Table 3. Effect of Circulating Liquid Velocity (u_{LR}) on the Volumetric Mass Transfer Coefficient ($k_La'_T$)*

u_{GR} (m/s)	u_{LR} (m/s)	η_{eff} (Pa · s)	$1 + A_d/A_r$	$k_La'_T$ (s^{-1})	$k_La'_T \cdot (1 + A_d/A_r)$ (s^{-1})
0.159	0.	0.09	1.	7.36	7.36
	0.038		1.111	5.93	6.59
	0.09		1.25	5.77	7.21
	0.16		1.444	5.30	7.65
0.043	0.	0.45	1.	1.30	1.30
	0.01		1.111	1.206	1.34
	0.028		1.25	1.04	1.30
	0.05		1.444	0.886	1.28

*System: Hercules 7H3SXF CMC/water solution phase oxygen diffusivity (D_L) in the sucrose solution (see Table 2). In order to develop a more generalized k_La_T correlation for viscous liquids of both Newtonian and non-Newtonian rheological behavior for BC and ECL-AL fermentors, we introduced into Eq. 26, which does not incorporate physicochemical property parameters other than the effective viscosity, the effects of liquid-phase diffusivity, mass density and interfacial tension as reported by Nakanoh and Yoshida (1980) and as listed in Table 1. The resulting generalized correlation is

$$k_La_T = (0.5)(10^{-2})u_{GR}^{0.52}D_L^{0.5}\rho_L^{1.03}[1 + (A_d/A_r)]^{-0.85} \cdot (\eta_{\text{eff}})^{-0.89}\sigma_L^{-0.75} \quad (27)$$

A parity plot of Eq. 27 is shown on Figure 8. As indicated there, Eq. 27 predicts k_La_T within $\pm 13\%$ for both bubble column ($A_d/A_r = 0$) and ECL-AL contactors containing either a viscous Newtonian liquid (51.8 wt. % sucrose solution) or one of a large number of types of CMC of different rheological characteristics. The correlation given by Eq. 27 was tested for the following ranges of experimental conditions:

$0.03 \le u_{GR} \le 0.26$ m/s

$0 \le A_d/A_r \le 0.444$

$0.02 \le \eta_{\text{eff}} \le 0.5$ Pa · s (for $\beta = 5{,}000$ m^{-1} in Eq. 3)

$0.33 \le D_L \cdot 10^9 \le 2.53$ m^2/s

$1{,}003 \le \rho_L \le 1{,}240$ kg/m^3

$59 \le \sigma_L \cdot 10^3 \le 79$ N/m

Different types of CMC have been employed in previous works for k_La measurements. Some of them are presented in Table 2 together with their physical properties. At the same CMC concentration, the consistency and flow behavior indices change significantly from type to type, depending upon the degree of polymerization. Considering the strong influence of η_{eff} on k_La (Eq. 27), comparison of k_La values measured in solutions of different types of CMC must take into consideration the differences in effective viscosity and not merely be based on the CMC weight percent as frequently has been done previously (e.g., Figure 13 in Deckwer et al., 1982).

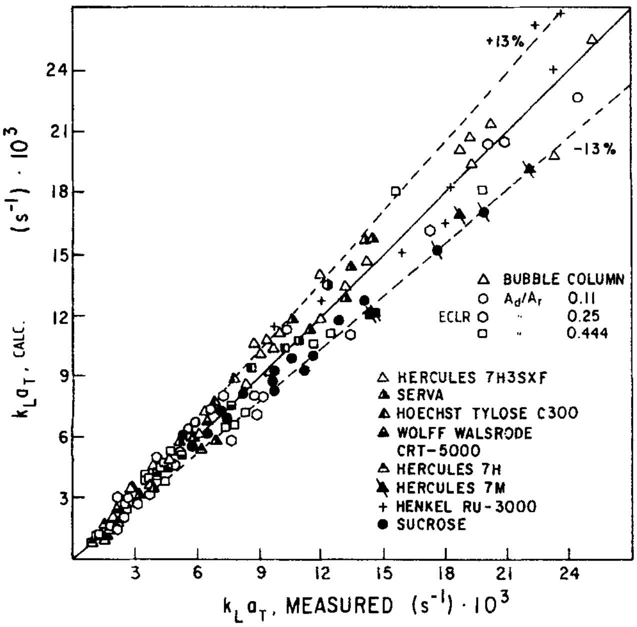

Figure 8. Parity plot of the generalized, k_La_T correlation, Eq. 27, for viscous liquids in BC and ECL-AL fermentors.

Predicted volumetric mass transfer coefficients (Eq. 27) for the BC case agree well with the findings of Deckwer et al. (1982), i.e., compare the values of the exponents on u_{GR} and η_{eff} given in Table 1 for the k_La function of Deckwer et al. (1982) with the corresponding values given in Eq. 27. Deckwer et al. (1982) also reported strong discrepancies between their results and those of Nakanoh and Yoshida (1980). König (1980) measured volumetric mass transfer coefficients during *Penicillium chrysogenum* fermentations in a 0.2 m i.d. bubble column of 2.10 m height and at gas superficial velocities of 0.04–0.05 m/s. The reported k_La values and rheological properties were used by Deckwer et al. (1982) for comparison. A good agreement between predicted (Deckwer et al., 1982) and measured (König, 1980) k_La values was reported by the former workers. Predicted mass transfer coefficients using the correlation of Godbole et al. (1984), given in Table 1, are lower by up to 50% than our experimental k_La data for the BC mode. This difference most likely is the result of the different flow regimes in each of these works. Equation 27 also predicts the k_La_T data of Onken and Weiland (1983) measured in 51.8 wt. % sucrose solution within $\pm 10\%$.

Table 4 compares values of k_La for the BC mode of operation computed from our correlation (Eq. 27 with $A_d/A_r = 0$) and from Eq. 4 recently presented by Schumpe and Deckwer (1987) over the range $0.023 \le \eta_{\text{eff}} \le 0.34$ Pa · s and for $\mu = 0.019$ Pa · s. In Table 4, the absolute or effective viscosities of the various aqueous solutions increase in the order: solution number 4, 1, 2, 3. For both Newtonian and non-Newtonian viscous liquids of relatively low viscosity (solutions 4 and 1), the Schumpe-Deckwer (1987) correlation yields values of k_La that are up to 22% lower than those predicted from Eq. 27; the discrepancy increases with increasing u_G (hence, increasing shear rate, see Eq. 3). With more viscous non-Newtonian CMC solutions (numbers 2 and 3 in Table 4), the discrepancies between these two correlations are much less, particularly at $u_G \ge 0.1$ m/s where the differences in the computed values of k_La are less than 9%. In the case of the CMC solutions, these differences largely are due to the different dependencies of k_La upon effective viscosity and interfacial tension in Eqs. 4 and 27, as determined by regression analysis in both cases, which discrepancies in turn appear to rise mainly from the different values of β used to estimate the shear rate from Eq. 3. We used $\beta = 5{,}000$ m^{-1}, as in the original work of Nishikawa et al. (1977), whereas Schumpe and Deckwer (1987) used $\beta = 2{,}800$ m^{-1}. They obtained the latter value by allowing β to be another adjustable parameter to be fitted by regression analysis of their data; $\beta = 2{,}800$ m^{-1} gave the best overall fit ($\Sigma\chi^2$ 6.2). They also noted that if they had used $\beta = 5{,}000$ m^{-1}, the goodness of fit of their data would not have changed appreciably, i.e., $\Sigma\chi^2 = 7.8$ (but, of course, the values of the fitted exponents on the η_{eff}, σ, etc. terms would be different from those given in Eq. 5).

Bello et al. (1985a,b) measured k_La_T in tap water and aqueous salt solution (0.4 kmol/m^3 NaCl) using an ECL-AL of

Table 4. Bubble Column Volumetric Mass Transfer Coefficients (k_La) Computed Using Eq. 27 (this work) or Eq. 4 (Schumpe and Deckwer, 1987*

Solution No.	K(Pa · s^n); n	u_G (m/s)	k_La (s^{-1}) × 10^3 Eq. 27 β = 5,000 m^{-1} A	Eq. 4 β = 2,800 m^{-1} B	% Deviation (B-A)(100)/A
1	0.21; 0.68	0.05	8.38	7.39	−11.8
		0.10	14.6	12.1	−17.1
		0.20	25.6	19.9	−22.3
2	2.03; 0.57	0.50	1.91	2.22	+16.2
		0.10	3.57	3.82	+7.0
		0.20	6.68	6.62	−0.9
3	9.23; 0.40	0.05	1.14	1.38	+21.2
		0.10	2.38	2.59	+8.8
		0.20	4.93	4.86	−1.4
4	51.8 wt. %	0.05	6.79	5.86	−13.7
	Sucrose	0.10	9.74	8.23	−15.5
	(Newtonian)	0.02	14.0	11.6	−17.1

*For all cases, D = 0.15 m; for all CMC solutions; D_L = 2.3 (10^{-9}) m^2/s, ρ_L = 1,005 kg/m^3 and σ_L = 75.5 (10^{-3}) N/m; 5.18 wt. % sucrose solution properties as given in Table 2

essentially the same geometry as in this work. They proposed a mass transfer coefficient correlation (see Table 1) that has the same type of functionality as Eq. 27, but which has very different values of the exponents on particular terms. The predicted and measured k_La_T values of Bello et al. (1985a,b) are at least an order of magnitude higher than in our viscous systems over the same range of u_{GR}. Therefore, direct comparison is not possible. As a paradox, introducing the value of water viscosity ($\mu \approx 0.001$ Pa · s) in the correlation of Nakanoh and Yoshida (1980), see Figure 7, leads to

$$k_La_T \approx 0.799\, u_G \cdot (1 - \epsilon_{GT}) \tag{28}$$

which is similar to the correlation of Bello et al. (1985a) for the BC mode. Thus the correlation of Nakanoh and Yoshida (1980), obtained in low viscosity solutions, cannot be recommended for k_La prediction for highly viscous fluids.

Conclusions

Hydrodynamic and mass transfer characteristics have been studied in ECL-AL fermentors and a BC with viscous non-Newtonian liquids (various CMC solutions) simulating mycelial fermentation broth, and with a viscous Newtonian liquid. The geometric parameter (A_d/A_r) and the riser gas superficial velocity (u_{GR}) were found to have the most significant affects on the mass transfer parameters, i.e., ϵ_{GR}, ϵ_{GD}, and k_La_T. The effective viscosity (η_{eff}) strongly influences k_La_T, but only weakly affects ϵ_{GR} and ϵ_{GD}. The circulating liquid superficial velocity (u_{LR}), which is dependent on u_{GR}, A_d/A_r and η_{eff}, has only a weak effect on the riser gas holdup and mass transfer coefficient. The average air bubble diameter in the downcomer of an ECL-AL fermentor is small, its rise velocity in highly viscous liquids is negligible, and hence, there is practically no slip between the phases in the downcomer. The gas holdup in the downcomer also is relatively small. Thus, the downcomer of an ECL-AL fermentor can be considered as a "dead" zone with respect to mass transfer.

Correlations of both ϵ_{GR} (Eq. 22) and k_La_T (Eq. 27) with bioreactor geometry (A_d/A_r), gas superficial velocity and liquid phase physical properties (viscosity for both ϵ_{GR} and k_La_T, plus diffusivity, density and interfacial tension for k_La_T) have been presented. The correlations are valid for both the bubble column ($A_d/A_r = 0$) and the ECL-AL ($0.11 \le A_d/A_r \le 0.444$) modes of operation. A limited amount of data suggest that the k_La_T correlation also is valid for highly-viscous Newtonian liquid systems. Applied to the bubble column case, the new correlations agree well with some of the results of previous workers, including measurements in real fermentation broths, for similar reactor geometry and gas-liquid flow pattern. The dispersion height (H_D) was not varied in this work, nor have there been previous systematic studies of the effect of H_D on ECL-AL mass transfer and hydrodynamics. Therefore, the effect of H_D on scale-up remains to be determined.

The mass transfer coefficient (k_La_T) decreases with increasing A_d/A_r, such that the bubble column mode of operation will give a higher oxygen transfer rate than an ECL-AL type of fermentor. On the other hand, due to the induced macro-scale liquid circulation in an ECL-AL fermentor, it has the potential for more rapid liquid-phase mixing (uniformity of substrate concentration, pH, temperature, etc.) than the corresponding BC. The relative circulating liquid velocities for the fermentor of Figure 1 at $A_d/A_r = 0$ (BC), 0.25 and 0.444 are 0., 0.573 and 1.0, respectively (Popovic and Robinson, 1988), whereas from Eq. 27 the relative k_La_T values are 1.0, 0.827, and 0.732, respectively. Therefore, the preferred ECL-AL design to achieve acceptable values of both the liquid-phase mixing time and the overall oxygen transfer coefficient in viscous, non-Newtonian fermentation broth may be one in which A_d/A_r is relatively small, e.g., $0.1 \le A_d/A_r \le 0.25$. Definitive mixing time studies in BC and ECL-AL fermentors for viscous non-Newtonian systems over a broad range of viscosity must be done in order to verify this tentative conclusion.

Acknowledgment

The authors gratefully acknowledge financial support from the Fermentation Technology Development and the Biotechnology Programmes of the National Research Council of Canada. M. Franke and

J. Huibers assisted at various times with experimental measurements and calculation.

Notation

a = specific interfacial area per unit dispersion volume, m^{-1}
a' = specific interfacial area per unit liquid volume, m^{-1}
a_1, a_2 = exponents in Eqs. 1 and 2
A_d = downcomer cross-sectional area, m^2
A_r = riser cross-sectional area, m^2
A_1, A_2 = proportionality constants in Eqs. 1 and 2
b_1, b_2 = exponents in Eqs. 1 and 2
c_2 = exponent in Eq. 2
C = concentration, $kmol/m^3$
d = bubble diameter, m
d_B = Sauter mean bubble diameter, $d_B = \Sigma_i n_i d_i^3 / \Sigma_i n_i d_i^2$, m
d_{hole} = sparger hole diameter, m
d_1, d_2 = exponents in Eqs. 1 and 2
D = column diameter, m
D_L = liquid-phase molecular diffusivity, m^2/s
e_1, e_2 = exponents in Eqs. 1 and 2
f_1, f_2 = exponents in Eqs. 1 and 2
F = interfacial area effective for mass transfer, m^2
G_1 = holdup function given by Eq. 1, dimensionless
G_2 = mass transfer coefficient function given by Eq. 2, s^{-1}
g_1, g_2 = exponents in Eqs. 1 and 2
g = gravitational constant, m/s^2
h_1, h_2 = exponents in Eqs. 1 and 2
H_D = dispersion height, m
H_L = gas-free liquid height, m
k_L = liquid-phase mass transfer coefficient, m/s
$k_L a$ = liquid phase volumetric mass transfer coefficient based on dispersion volume, s^{-1}
$k_L a'$ = liquid-phase volumetric mass transfer coefficient based on liquid volume, s^{-1}
K = power law consistency index, $Pa \cdot s^n$
n = power law flow behaviour index, dimensionless
t = time, s
u = superficial velocity, m/s
V = volume, m^3

Greek letters

β = proportionality constant in Eq. 3, m^{-1}
χ = relative error
ϵ = fractional gas holdup, dimensionless
η = non-Newtonian liquid-phase viscosity, $Pa \cdot s$
$\dot{\gamma}$ = shear rate, s^{-1}
μ = Newtonian liquid-phase viscosity, $Pa \cdot s$
ρ_L = liquid-phase mass density, kg/m^3
σ = interfacial tension with respect to air, N/m
τ = shear stress, Pa

Subscripts

B = bubble
cal = calculated
eff = effective (for viscosity)
d, D = downcomer, dispersion
G = gas
i = bubble size class
L = liquid
meas = measured
o = initial condition
r, R = riser
T = total

Superscripts

$*$ = value in equilibrium with bulk-phase property of the other phase

Literature Cited

Akita, K., and F. Yoshida, "Gas Holdup and Volumetric Mass Transfer Coefficient in Bubble Columns," *Ind. Eng. Chem. Process Des. Dev.*, **12**, 76 (1973).

Akita, K., and F. Yoshida, "Bubble Size, Interfacial Area and Liquid-Phase Mass Transfer Coefficient in Bubble Columns," *Ind. Eng. Chem. Process Des. Dev.*, **13**, 84 (1974).

Allen, D. G., M. Popovic, and C. W. Robinson, "Pneumatically Agitated Bioreactor Performance Characteristics: Are Predictions Based on Simulation Studies Applicable to Real Mycelial Fermentations," Eur. Cong. on Biotechnology, Amsterdam, Netherland (June 14–19, 1987).

Bello, R. A., "A Characterization Study of Airlift Contactors for Application to Fermentors," Ph.D. Thesis, Univ. of Waterloo, Canada (1981).

Bello, R. A., C. W. Robinson, and M. Moo-Young, "Liquid Circulation and Mixing Characteristics of Airlift Contactors," *Can. J. Chem. Eng.*, **62**, 573 (1984).

Bello, R. A., C. W. Robinson, and M. Moo-Young, "Gas Holdup and Overall Volumetric Oxygen Transfer Coefficient in Airlift Contactors," *Biotechnol. Bioeng.*, **27**, 369 (1985a).

Bello, R. A., C. W. Robinson, and M. Moo-Young, "Prediction of the Volumetric Mass Transfer Coefficient in Pneumatic Contactors," *Chem. Eng. Sci.*, **40**, 53 (1985b).

Buchholz, H., R. Buchholz, J. Lücke, and K. Schügerl, "Bubble Swarm Behavior and Gas Absorption in Non-Newtonian Fluids in Sparged Columns," *Chem. Eng. Sci.*, **33**, 1061 (1978).

Chakravarty, M., S. Begum, H. D. Singh, J. N. Baruah, and M. S. Jyengar, "Gas Holdup Distribution in a Gas-Lift Column," *Biotechnol. Bioeng. Symp.*, **4**, 363 (1973).

Deckwer, W. D., K. Nguyen-Tien, A. Schumpe, and Y. Serpemen, "Oxygen Mass Transfer into Aerated CMC Solutions in a Bubble Column," *Biotechnol. Bioeng.*, **24**, 461 (1982).

Devine, W. D., Y. T. Shah, and B. J. Morsi, "Liquid Phase Axial Mixing in a Bubble Column with Viscous non-Newtonian Liquids," *Can. J. Chem. Eng.*, **63**, 195 (1985).

Godbole, S. P., M. F. Honath, and Y. T. Shah, "Holdup Structure in Highly Viscous Newtonian and Non-Newtonian Liquids in Bubble Columns," *Chem. Eng. Comm.*, **16**, 119 (1982).

Godbole, S. P., A. Schumpe, Y. T. Shah, and M. L. Carr, "Hydrodynamics and Mass Transfer in non-Newtonian Solutions in a Bubble Column," *AIChE J.*, **30**, 213 (1984).

Hikita, H., S. Asai, and Y. Azuma, "Solubility and Diffusivity of Oxygen in Aqueous Sucrose Solutions," *Can. J. Chem. Eng.*, **56**, 371 (1978).

König, B., "Untersuchungen zur Penicillinproduktion in Blasensäulen," Ph.D. Thesis, Univ. of Hannover, FRG (1980).

Kristiansen, B., "Trends in Fermentor Design," *Chem. Ind.*, London, **21**, 787 (1978).

Lin, C. H., B. S. Fang, C. S. Wu, H. Y. Fang, T. F. Kuo, and C. Y. Hu, "Oxygen Transfer and Mixing in a Tower Cycling Fermentor," *Biotechnol. Bioeng.*, **23**, 1557 (1976).

Nakanoh, M., and F. Yoshida, "Gas Absorption by Newtonian and Non-Newtonian Liquids in a Bubble Column," *Ind. Eng. Chem. Process Des. Dev.*, **19**, 190 (1980).

Navari, R. M., J. L. Gainer, and K. R. Hall, "A Predictive Theory for Diffusion in Polymer and Protein Solutions," *AIChE J.*, **17**, 1028 (1971).

Nishikawa, M., H. Kato, and K. Hashimoto, "Heat Transfer in Aerated Tower Filled with Non-Newtonian Liquid," *Ind. Eng. Chem. Process Des. Dev.*, **16**, 133 (1977).

Onken, U., and P. Weiland, "Hydrodynamic and Mass Transfer in an Airlift Loop Fermentor," *Eur. J. Appl. Microbiol. Biotechnol.*, **10**, 31 (1980).

Onken, U., and P. Weiland, "Airlift Fermentors: Construction, Behavior and Uses," *Adv. Biotechnol. Proc.*, **1**, 67 (1983).

Patwari, A. N., "Hydrodynamics and Mass Transfer in Three Phase Fluidized Beds with Highly Viscous Liquids," Ph.D. Thesis, Univ. of Oldenburg, FRG (1983).

Popovic, M., and C. W. Robinson, "Estimation of Some Important Design Parameters for Non-Newtonian Liquids in Pneumatically-Agitated Fermentors," *Proc. Can. Chem. Eng. Conf.*, Quebec, 258 (Oct., 1984).

Popovic, M., and C. W. Robinson, "The Specific Interfacial Area in External-Circulation-Loop Airlifts and a Bubble Column: I. Aqueous Sodium Sulfite Solution, *Chem. Eng. Sci.*, **42**, 2811 (1987a).

Popovic, M., and C. W. Robinson, "The Specific Interfacial Area in External-Circulation-Loop Airlifts and Bubble Column: II. CMC/ Sulfite Solution," *Chem. Eng. Sci.*, **42**, 2825 (1987b).

Popovic, M., and C. W. Robinson, "Circulating Liquid Velocity in External-Circulation-Loop Air-Lift Fermentors," *Biotechnol. Bioeng.*, (in press, 1988).

Schumpe, A., "Die Chemische Bestimmung von Phasengrenzflächen in Blasensäulen bei Uneinheitlichen Blasengrössen," Ph.D. Thesis, Univ. of Hannover, FRG (1981).

Schumpe, A., and W.-D. Deckwer, "Gas Holdups, Specific Interfacial Areas and Mass Transfer Coefficients of Aerated Carboxymethyl Cellulose Solutions in a Bubble Column," *Ind. Eng. Chem. Process Des. Dev.*, **21**, 706 (1982).

Schumpe, A., and W.-D. Deckwer, "Viscous Media in Tower Bioreactors: Hydrodynamic Characteristics and Mass Transfer Properties," *Bioprocess Eng.*, **2**, 79 (1987).

Shah, Y. T., B. G. Kelkar, S. P. Godbole, and W.-D. Deckwer, "Design Parameters Estimations for Bubble Column Reactors," *AIChE J.*, **30**, 353 (1982).

Stein, W. A., and H. Schäfer, "Aeration of Viscous Liquids with a Downward Directed Two-Fluid Nozzle," *Ger. Chem. Eng.*, **6**, 91 (1983).

Voigt, J., V. Hecht, and K. Schügerl, "Absorption of Oxygen in Countercurrent Multistage Bubble Columns: II. Aqueous Solutions with High Viscosity," *Chem. Eng. Sci.*, **35**, 1317 (1980).

Yagi, H., and F. Yoshida, "Gas Absorption by Newtonian and Non-Newtonian Fluids in Sparged Agitated Vessel," *Ind. Eng. Chem. Process Des. Dev.*, **14**, 488 (1975).

Manuscript received Aug. 12, 1987 and revision received Oct. 14, 1988.