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Gas Holdup in Three-Phase Immobilized-Cell Bioreactors

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ABSTRACT

A number of studies in the published literature deal with gas holdup in three-phase reactors. However, very few address the cases in which the solid density approaches that of the liquid phases and in which low gas velocities are involved. These conditions are commonly encountered in immobilized-cell bubble columns and in fluidized-bed bioreactors. This paper reports the effect of gas and liquid velocity upon gas holdup and bed expansion in fluidized-bed bioreactors.

For liquid-fluidization of low-density alginate beads in the absence of gas, the terminal sedimentation velocity (v_T) of the particles is a constant, and expansion of the bed follows Richardson and Zaki's correlation. In the presence of gas, however, the apparent terminal sedimentation velocity value is affected by the velocity of the gas and liquid phases. For gas velocities above a minimum value, the calculated value of v_T depends on liquid velocity only, and a constant bed expansion was observed for a range of gas and liquid flow rates. For the gas–liquid interactions, a modified drift-flux model was found to be valid. For superficial gas velocities between 5 and 17 cm/min, the modified drift-flux velocity was observed to be a function of gas velocity, suggesting the prevalence of a coalescence regime.

Index Entries: Hydrodynamics; drift-flux; wake-model; multiphase, fluidized bed.

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NOMENCLATURE

k	ratio of fractions of wake and gas phases
n	exponent in Richardson and Zaki's equation
u_B	rise velocity of bubbles, cm/min
$u_{B\infty}$	terminal rise velocity of bubbles, cm/min
u_g	superficial gas velocity, cm/min
u_L	superficial liquid velocity, cm/min
$v_{g\text{-}L}$	drift-flux velocity, cm/min
v_T	sedimentation velocity of the solids, cm/min
v_T^*	a pseudo settling velocity
x	ratio of fractions of solids in the wake and in the liquid
α	exponent in the drift-flux model
ϵ_g	gas holdup
ϵ_L	liquid holdup
ϵ_s	solid holdup

INTRODUCTION

Three-phase fluidized-bed reactors with immobilized biocatalysts have recently been explored for a number of bioprocesses, including ethanol fermentation, waste treatment, and biodegradation (*1–7*). In many of these systems, solid particles containing the biocatalysts are fluidized by liquid, resulting in a two-phase system. Usually there is also a gas phase present, either as a substrate, like oxygen, or a product, such as CO_2. The gas phase changes the hydrodynamic behavior in the now three-phase reactor. Two important hydrodynamic variables affected are the gas holdup and the backmixing caused by the rising bubbles. For the proper design and scaleup of such systems, a knowledge of the local values of holdup of the gas and solid phases, together with mixing in the different phases, is essential. This paper deals with the prediction and correlation of gas holdup in three-phase systems. The knowledge of holdup is important because the solid holdup is the biocatalyst concentration, the gas holdup is the void unavailable for reaction, and the gas holdup and bubble size determine the surface area for mass transfer. Holdups are known to be affected by the gas and liquid velocities and properties; the size, amount, and density of solids; and the sparger, if present.

Hydrodynamic correlations on these systems are tentative (*8*), especially since most data were collected using high-density particles (glass and metal, with a minority using plexiglas™ (specific gravity ~1.2), and virtually none using particle densities near that of the liquid (as is common for bioprocesses). In this approach to the complex three-phase system, we will first look at the liquid–solid interaction and then the gas–liquid interactions. Direct gas–solid interactions (not mediated by the liquid) are presumed to be unimportant at this stage.

THEORETICAL BACKGROUND

The fluidization of solid particles by liquids has been a subject of numerous studies. Under conditions of homogeneous liquid–solid fluidization, the volume fraction or holdup of the liquid phase (ϵ_L) is related to the superficial liquid velocity (u_L) by the following equation proposed by Richardson and Zaki (*9*):

$$u_L = v_T \cdot \epsilon_L{}^n \quad (1)$$

where v_T is terminal settling velocity of the particles and the exponent n depends on the Reynolds number in the reactor column. The value of n has been reported to range between 2.4 and 4.65, although values as high as 20 have been reported; n is also reported to decrease with an increasing Reynolds number in a two-phase system (*9*).

When gas is introduced in the liquid-fluidized bed of solid particles, the bed may expand or contract, depending on the relative sizes of the bubbles and the particles, and on the wettability of the solid surface. Although many attempts have been made to explain this phenomenon, "wake models" (*10–14*) have been the most successful. These models consider the three-phase system as consisting of a gas phase (bubbles), a phase consisting of solid particles and liquid transported in the wake of these bubbles, and a third phase, consisting of the remaining liquid-fluidized solids. A generalized model proposed by Bhatia and Epstein (*15*) results in the following expression for the holdup or volume fraction of the liquid phase, (ϵ_L):

$$\epsilon_L = k\,\epsilon_g\,(1 - x) + \{1 - \epsilon_g(1 + k - k \cdot x)\} \cdot [(u_L - u_g\,k\,(1 - x))\,/\,v_T\{1 - \epsilon_g\,(1 + k)\}]^{\{1/n\}} \quad (2)$$

where k is the ratio of fractions of reactor volume occupied by bubble-wakes and by the gas holdup, (ϵ_g) and x is the ratio of fractions of solids in the bubble-wakes and in the remaining phase containing liquid-fluidized solids. For k and x, a number of correlations relating these important variables to the velocities of the gas (u_g) and liquid (u_L) phases have been proposed in the literature. A set of commonly used correlations proposed by El-Temtamy and Epstein (*12*) is given below:

$$k = (0.61 + 0.037\,/\,(\epsilon_g + 0.013)) \cdot (\epsilon_g + \epsilon_L)^3 \quad (3)$$

$$\begin{aligned} x &= 1.0 - 0.877\,v_T\,/\,(u_g/\epsilon_g - u_L/\epsilon_L) && \text{for } 0 < v_T\,/\,(u_g/\epsilon_g - u_L/\epsilon_L) < 1.14 \\ x &= 0.0 && \text{for } 1.14 < v_T\,/\,(u_g/\epsilon_g - u_L/\epsilon_L) \end{aligned} \quad (4)$$

Equations (2–4) can be used to predict theholdup patterns in the reactor, provided appropriate correlations for the behavior of the solid–liquid and the gas–liquid systems are available. Frequently these correlations need to be determined for the chosen experimental system. For the solid-liquid part of the system, Richardson and Zaki's correlation (Eq. [1]) is

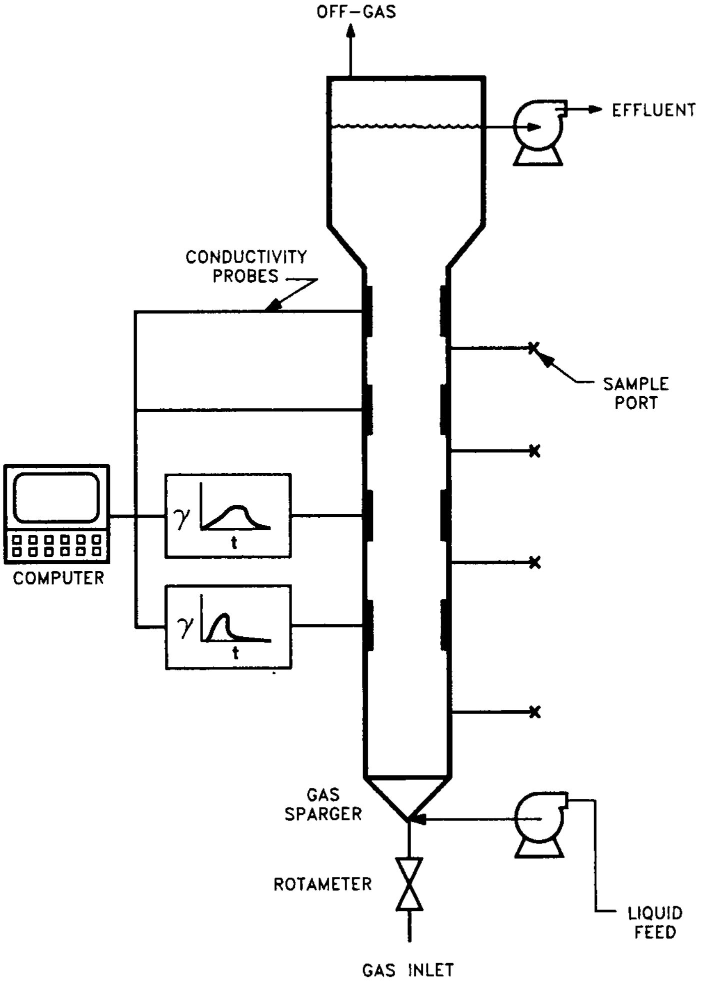

Fig. 1. Schematic diagram of the fluidized-bed reactor.

commonly used. There are reasons to believe that the parameter v_T is affected by the presence of gas in the system, and no satisfactory relation to account for this effect is available.

For the gas-liquid part of the system, a number of correlations have been used. Several authors have recently proposed the use of a drift-flux model suggested by Wallis (*16*) for this purpose. According to this model, the drift-flux velocity ($v_{g\text{-}L}$), which is proportional to the difference between the actual velocity of the gas phase (u_g/ϵ_g) and the total velocity in the combined two-phase gas-liquid system, can be related to the characteristic bubble-rise velocity ($u_{B\infty}$) in the reactor. For a two-phase system:

$$v_{g-L} = [u_g / \epsilon_g - u_L / \epsilon_L] \cdot \epsilon_g \cdot \epsilon_L = u_{B\infty} \cdot \epsilon_g \cdot (1 - \epsilon_g)^{\alpha}. \quad (5)$$

The value of α depends on the size of bubbles and varies between 0 and 2. For small values of ϵ_g, the term involving α can be neglected. This model has been shown to be applicable to two-phase systems with changing hydrodynamic regimes (*17*). The effect of the hydrodynamic regimes is accounted for by $u_{B\infty}$, which depends on sparger characteristics, gas flow rate, and the liquid hydrodynamics. These dependencies vary in different regimes of operation. The presence of solids also influences the value of this parameter, but no data are available for this effect.

MATERIALS AND METHODS

Experiments were conducted in a 7.64-cm id, 6-L, columnar, fluidized-bed reactor fitted with a 7.64-cm id, medium glass fritt at the bottom, through which a metered air-stream was introduced. The liquid (0.05*M* KCl) was introduced through four radial inlet ports located directly above the sparger plate. In this reactor, a known volume of $\varkappa$-carrageenan beads (4% gel, 3% Fe_2O_3, mean diameter 0.125 cm) were fluidized with liquid and/or gas. The conductivities of the electrolyte and the suspended gel beds have been measured (*18*) to be almost equal (within 2%) and were considered identical. The holdup of gas phase was measured with the help of several conductivity probes located in the column. Solid holdup was measured by monitoring the height of the bed. In previous experiments (*19*), there was the formation of a stable bubble size of about 4 mm in diameter for all flow rates. The bed/particle ratios of 60 and bed/bubble ratios of 20 suggest that wall effects should be minimal; but further experiments may be necessary to confirm this conclusion. Detailed experimental procedures can be found elsewhere (*18,19*), along with some of the raw data. More data and the theoretical interpretation and correlations are presented here. A schematic diagram is shown in Fig. 1.

RESULTS AND DISCUSSION

A number of measurements of gas and solid holdup were made at different flow rates of gas (0–17 cm/min) and liquid (0–17 cm/min). The data from the three-phase system are presented in Table 1.

Applicability of Richardson and Zaki's Correlation

The results of liquid-fluidization experiments conducted in the absence of gas are plotted in Fig. 2. With increasing liquid velocities, the bed gradually expanded, resulting in an increase in the value of liquid holdup in the fluidized bed. From Fig. 2, the applicability of Richardson and Zaki's equation is evident. For the $\varkappa$-carrageenan beads, a linear least-squares estimation gave the following relation between liquid holdup in the two phase solid-liquid system and liquid velocity

$$u_L = 52.24\,(\epsilon_L)^{3.43}. \tag{6}$$

Table 1
Experimental and Predicted Holdup Values for the Three-Phase FBR

Experimental values					Predicted values		
u_L, cm/min	u_g, cm/min	ϵ_s	ϵ_g	ϵ_L	ϵ_s	ϵ_g	ϵ_L
			Volume of beads = 1000 mL				
11.05	7.3	0.16	0.011	0.83	0.173	0.0015	0.817
12.62	7.3	0.16	0.013	0.83	0.172	0.0106	0.817
7.13	7.3	0.18	0.014	0.81	0.178	0.0146	0.807
12.22	7.3	0.18	0.009	0.82	0.173	0.0108	0.817
10.91	7.3	0.18	0.011	0.81	0.173	0.0116	0.815
9.67	7.3	0.18	0.012	0.80	0.174	0.0124	0.813
14.14	7.3	0.17	0.010	0.83	0.172	0.0098	0.819
14.03	7.3	0.17	0.011	0.82	0.172	0.0098	0.819
16.8	7.3	0.16	0.011	0.83	0.164	0.009	0.827
12.58	12	0.17	0.012	0.82	0.171	0.014	0.815
11.09	12	0.17	0.014	0.82	0.172	0.015	0.813
5.27	12	0.17	0.015	0.81	0.185	0.020	0.795
15.49	12	0.17	0.012	0.82	0.170	0.0126	0.817
8.73	12	0.18	0.016	0.81	0.175	0.0168	0.809
7.2	12	0.17	0.018	0.81	0.177	0.0182	0.805
11.89	13.1	0.17	0.013	0.81	0.172	0.015	0.813
11.67	10.8	0.17	0.012	0.81	0.172	0.014	0.814
11.67	8.9	0.17	0.010	0.82	0.173	0.0125	0.815
17.67	7.3	0.17	0.009	0.83	0.164	0.0086	0.828
16.76	5.7	0.17	0.008	0.83	0.164	0.0075	0.828
11.67	17.1	0.18	0.020	0.80	0.171	0.0173	0.812
11.52	14.8	0.18	0.017	0.81	0.172	0.0163	0.812
11.63	17.1	0.17	0.018	0.81	0.171	0.0173	0.812
16.78	17.1	0.17	0.019	0.82	0.169	0.0146	0.817
17.23	14.8	0.17	0.018	0.82	0.169	0.0134	0.818
16.78	13.1	0.17	0.016	0.81	0.169	0.0127	0.818
15.58	13.1	0.17	0.016	0.81	0.171	0.0132	0.817
			Volume of beads = 800 mL				
16.94	17.1	0.20	0.017	0.78	0.169	0.0145	0.817
16.91	14.8	0.20	0.018	0.78	0.169	0.0135	0.817
16.91	13.1	0.21	0.016	0.78	0.169	0.0126	0.818
16.91	10.9	0.20	0.012	0.79	0.170	0.0113	0.819
16.91	8.9	0.20	0.015	0.78	0.170	0.010	0.820
17.01	7.3	0.21	0.011	0.78	0.164	0.009	0.827
17.01	5.7	0.21	0.010	0.78	0.164	0.0074	0.829
11.42	17.1	0.20	0.018	0.78	0.171	0.0174	0.811
11.42	14.8	0.21	0.022	0.77	0.172	0.0164	0.812
12.0	13.1	0.23	0.016	0.76	0.172	0.015	0.813
12.0	10.8	0.22	0.015	0.77	0.172	0.014	0.814
12.0	8.9	0.22	0.017	0.76	0.172	0.012	0.815

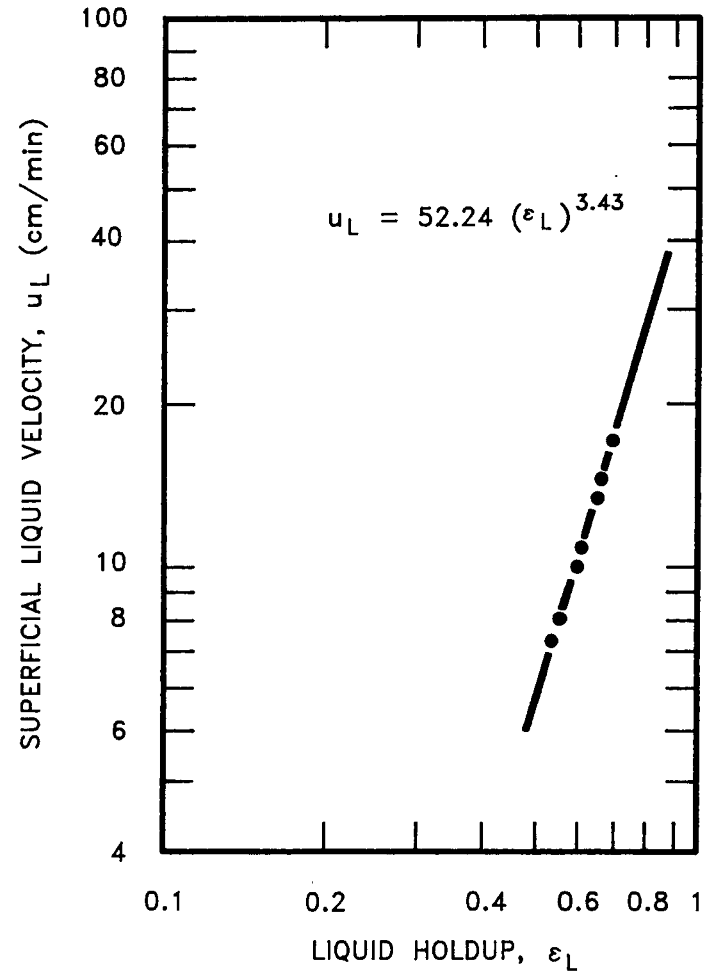

Fig. 2. Superficial liquid velocity vs liquid holdup in a two-phase solid–liquid FBR. Solid line represents data-fit by correlation of Richardson and Zaki.

Here, v_T was constant at 52 cm/min, which is reasonable for these particles. A value of n of 3.7 can be obtained from Richardson and Zaki's correlations using the experimental v_T. A key question is whether v_T will remain constant in the three-phase system. In the presence of gas, a modified equation may be written as

$$u_L / (1 - \epsilon_g) = v_T \cdot [\epsilon_L / (1 - \epsilon_g)]^n \tag{7}$$

This equation was formulated by considering the solids to be suspended only in the liquid-solid fraction (not in the gas fraction of the three-phase system), by neglecting the separation of the liquid–solid fraction into wake and nonwake portions, and by assuming the applicability of the Richardson and Zaki's correlation for this part. The equivalence of n in both Eqs. (6) and (7) is unknown; but as a first approximation, they were presumed

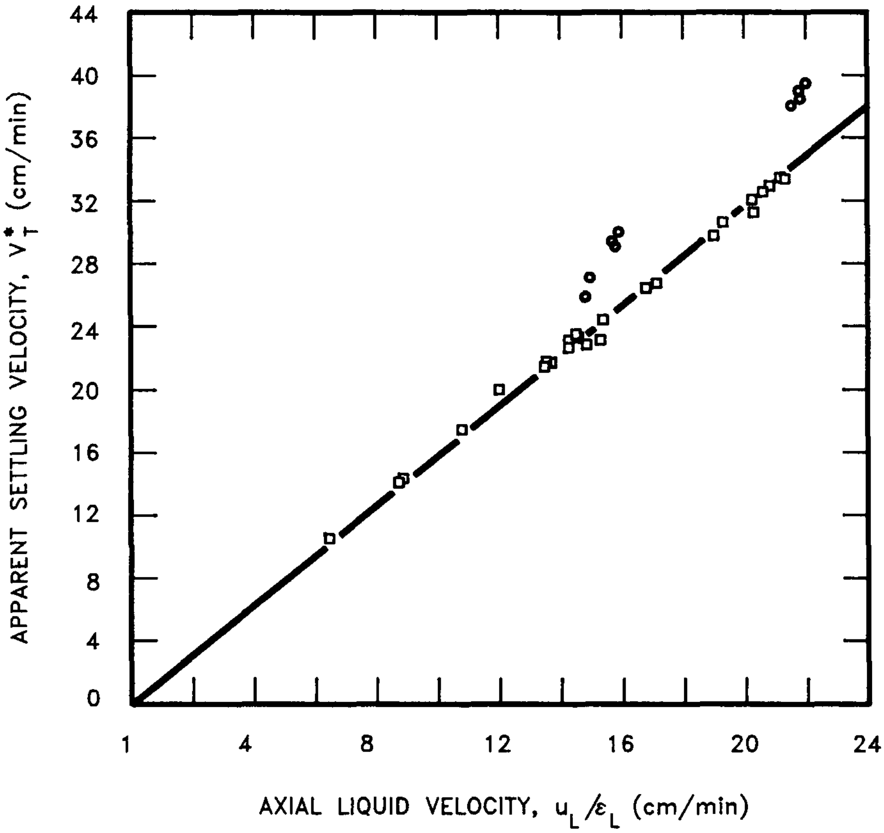

Fig. 3. Apparent particle sedimentation velocity, calculated from Eq. (7); vs liquid velocity in the three-phase FBR. O: data from lower loading experiments, 810 mL; X: data from higher loading experiments, 1000 mL (Table 1).

equal here. As suggested earlier, it is suspected that the terminal settling velocity of the particles (v_T) may be affected by the presence of gas (i.e., it is not constant). In order to study this effect, the values of v_T^*, the apparent settling velocity, were calculated from experimental data in Table 1 using Eq. (7); v_T becomes a fitted parameter. These values suggest that v_T^* has little dependence on the gas velocity. On the other hand, v_T^* depends linearly upon the liquid velocity (Fig. 3). The data for experiment numbers 159–184 can be correlated with the help of the following relation

$$v_T^* = 1.52\, u_L/\epsilon_L \tag{8}$$

This observed dependence of the apparent terminal settling velocity (v_T^*) on liquid velocity is surprising; but it clearly arises out of the experimental observation (Table 1) that the bed, in the presence of gas, expanded to an almost constant extent for the whole range of gas and liquid flow rates employed in this study. The reasons why this happened are not clear, although similar phenomena have been reported by others also for fluidization of low-density particles (*20*). For beads of plexiglas™ (specific gravity 1.17, Begovich (*20*) reported that the liquid holdup attained a constant value above a small gas velocity through the column, and was not affected by the liquid flow rate. This was not observed for the fluidization of high-density particles, such as glass (specific gravity 2.2) and lead, which are typically used in fluidization studies. It would appear that the presence of small amounts of gas in the liquid destabilizes the sedimentation of particles sufficiently so that any further changes in the gas flow rate are not important. Richardson and Zaki's correlation (as explicitly stated in Eq. [7]) is not valid for this three-phase system. However, if this equation is still used to analyze the data, an artifact is observed in which the apparent sedimentation velocity (v_T*) of the particles is linearly related to the liquid velocity (Fig 3). In that case, v_T* is a pseudoparameter used to fit the data.

In order to understand the phenomenon of constant liquid holdup in this three-phase system, one could look at the minimum fluidization velocity in a three-phase system. It has been reported that the minimum liquid-fluidization velocity is lowered by the presence of gas (*20*). As a result, a bed of liquid-fluidized solid particles expands with the introduction of gas (*20*). In the case of low-density alginate beads, Su (*21*) has reported minimum gas velocities. These values ranged from 0.4 to 2 cm/min. For the experiments reported in Table 1, the minimum gas velocities employed were >5 cm/min, which would be above the minimum fluidization velocities of gas alone for this three-phase system. Operation in a regime in which either the liquid or the gas flow is sufficient to fluidize the solid may indicate when the phenomenon of a constant bed expansion occurs. Perhaps an increase in liquid velocity lowers the energy imparted by the gas bubbles to the solids in a way such that the level of the bed expansion does not change. This hypothesis requires further scrutiny.

From Fig. 3, it appears that there is also a minor influence of particle loading upon the interactions between settling velocity and the gas and liquid flow rates. This too needs to be explored further.

Applicability of Drift-Flux Model

For the three-phase gas-liquid-solid fluidization, an expression for drift-flux velocity ($v_{g\text{-}L}$) was derived, in a manner similar to Eq. (5) for low ϵ_g, by accounting for the direct volume occupied by the solids.

$$v_{g\text{-}L} = [u_g / \epsilon_g - u_L / \epsilon_L]\, \epsilon_g\, \epsilon_L / (1 - \epsilon_s)^2 = u_{B\infty}\, \epsilon_g / (1 - \epsilon_s) \qquad (9)$$

In Fig. 4, the data from the three-phase, gas-liquid-solid fluidized-bed reactor are plotted as the modified drift-flux velocity, $u_g \cdot (1-\epsilon_s)$ or, equivalently, $u_{B\infty} \cdot \epsilon_g$, against the superficial gas velocity, u_g. For the gas and liquid velocities used in this work, the bubble-rise velocity increases with gas velocity in the system. The linear relation between these two terms suggests that the hydrodynamics is in the transition regime. In the bubbly flow as well as the churn-turbulent flow regimes, the characteristic bubble-

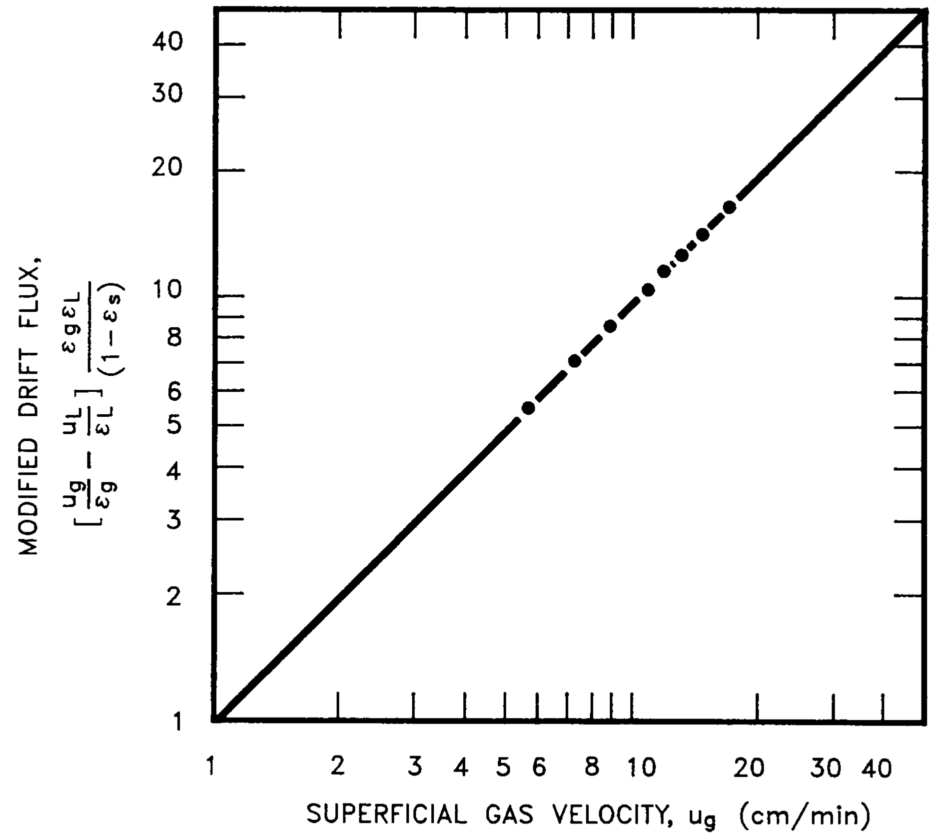

Fig. 4. Plot of modified drift-flux velocity, $v_{g\text{-}L}\cdot(1-\epsilon_s)$ calculated from Eq. (9) vs the superficial gas velocity, u_g. This plot uses all three-phase FDR data in Table 1.

rise velocity, $u_{B\infty}$ stays constant (*see* Eq. [5]); but for the transition or coalescence regime, it changes with gas velocity u_g/ϵ_g. Coalescence between bubbles dominates the hydrodynamics, resulting in the establishment of the observed stable bubble diameter. The following relation for the gas-liquid region was obtained (after rearrangement to eliminate terms, and considering $u_{B\infty}$ to be proportional to u_g/ϵ_g):

$$[u_g / \epsilon_g - u_L / \epsilon_L]\, \epsilon_g\, \epsilon_L / (1 - \epsilon_s) = 0.965\, u_g \tag{10}$$

Sufficient equations and parameters have been formulated to potentially characterize the system. Then Eqs. (8) and (10) and their constants (derived from the data) were used in the wake-model Eqs. (2–4) to calculate recursively the fractions of gas, liquid, and solid for the different experimental conditions. These calculated values are also presented in Table 1. The agreement between the measured and the calculated values is quite satisfactory, indicating that the methodology is able to characterize the data.

It can, therefore, be concluded that the drift-flux model of Wallis (*16*) can be used for the gas–liquid portion of the three-phase fluidized-bed reactors. However, attention must be paid to the hydrodynamic regime prevailing in the reactor, since the characteristic parameters will have different values in the different regimes. The presence of solid particles in the system can also have profound influence on the hydrodynamics in the column. This could change the characteristics of bubbles formed at the sparger as well as the coalescence between bubbles. Both of these affect the characteristic bubble-rise velocity (*21*). More experimental verification in the different regimes is recommended to expand the applicability of accurate hydrodynamic correlations.

CONCLUSIONS

The correlation and estimation of gas holdup in three-phase reactors has been explored. As expected for two-phase, liquid–solid fluidized beds, the Richardson and Zaki equation works very well. However, in gas–liquid fluidized beds with low-density solids, the experimental observation that the liquid holdup remains constant for a range of gas velocities has important ramifications on the correlations used. These include a linear dependence of the particle pseudosettling velocity on the liquid velocity. An argument for the underlying cause of this phenomenon has been presented, but requires further research.

The drift-flux model has been shown to correlate the experimental data very well. For the operating conditions of gas and liquid velocities used in the work, a coalescing regime was found to hold. The correlations of these types, when combined with the wake-model for three-phase fluidization, should allow an accurate prediction of the gas holdup in the presence of low-density solids, such as biocatalysts.

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