

# Particle Circulation and Oxygen Mass Transfer in an Immobilized Cell Bioreactor

JONATHAN F. DEAN AND COLIN WEBB\*

*Department of Chemical Engineering, U.M.I.S.T.,  
Manchester, M60 1QD, England*

Received August 4, 1986; Accepted March 9, 1987

## ABSTRACT

Two important considerations in the design of an aerobic particulate immobilized cell bioreactor are the provision of sufficient oxygen to maintain the desired metabolism of the immobilized organism, and the biomass holdup (which is proportional to the number of immobilized cell particles in the reactor).

The Circulating Bed Reactor, a reactor developed for use with those forms of immobilization that result in particles of essentially neutral buoyancy, operates with an expanded bed of circulating particles. The particle number density attainable in such a reactor has been found to be dependent upon the circulation cell aspect ratio, the individual particle properties, the static bed voidage of the particles, and the superficial gas velocity. The oxygen mass transfer characteristics have been found to be dependent upon the circulatory nature of the system, the particle (solids) holdup, the particle porosity, and the superficial gas velocity.

**Index Entries:** Particle circulation, in an immobilized cell bioreactor; oxygen mass transfer, in an immobilized cell bioreactor.

## NOMENCLATURE

$a$	Specific interfacial area for mass transfer.	$L^{-1}$
$C_L^*$	Equilibrium oxygen concentration in the liquid.	$ML^{-3}$
$C_L$	Bulk liquid oxygen concentration.	$ML^{-3}$

\* Author to whom all correspondence and reprint requests should be addressed.

$d_b$	Sauter mean bubble diameter.	L
$k_L$	Liquid film mass transfer coefficient.	$LT^{-1}$
$k_{La}$	Volumetric mass transfer coefficient.	$T^{-1}$
$m_p$	Mean biomass holdup within an immobilized cell particle.	M
$N_L$	Oxygen transfer rate based on liquid volume.	$ML^{-3} T^{-1}$
$N_p$	Number of immobilized cell particles within a bioreactor.	—
$N_R$	Oxygen transfer rate based on reactor volume.	$ML^{-3} T^{-1}$
$N_S$	Oxygen transfer rate based on solids volume.	$ML^{-3} T^{-1}$
$R_p$	Mean specific rate of reaction of immobilized biomass.	$T^{-1}$
$R_v$	Volumetric rate of reaction of a chemical reactor.	$ML^{-3} T^{-1}$
$U_{bs}$	Bubble rise velocity.	$LT^{-1}$
$U_g$	Superficial gas velocity.	$LT^{-1}$
$U_t$	Terminal velocity.	$LT^{-1}$
$V_p$	Particle volume.	$L^3$
$V_R$	Reactor volume.	$L^3$
$\epsilon_g$	Mean volumetric gas holdup.	—
$\epsilon_{sa}$	Apparent solids holdup.	—
$\epsilon_s$	Actual solids holdup.	—
$\phi$	Fraction of base area of circulating bed used for aeration.	—
$\psi$	Particle porosity.	—

## INTRODUCTION

The performance of a bioreactor is dependent on the volumetric rate of reaction ( $R_v$ ), which can be defined as the product of the net specific rate of reaction, ( $R_p$ ) and the amount of biomass in the reactor (the biomass holdup). With immobilized cell bioreactors the biomass holdup is largely independent of operational parameters such as dilution rate, and this leads to several novel process possibilities, including continuous operation at high throughput, continuous production of secondary metabolites, batch operation with large inoculum, and increased volumetric productivities.

There are several methods available for cell immobilization (1), which result in the formation of discrete particles. Consequently, there is some diversity in the size, shape, and density of such particles, and in the mode of operation of immobilized cell reactors (2).

Immobilization techniques being developed at UMIST rely on the natural accumulation of cells on or within inert support material, and include the porous Biomass Support Particles (BSPs) of Atkinson et al. (3). These consist of highly porous matrices within which there exists a relatively low shear environment compared to that outside the particles. The

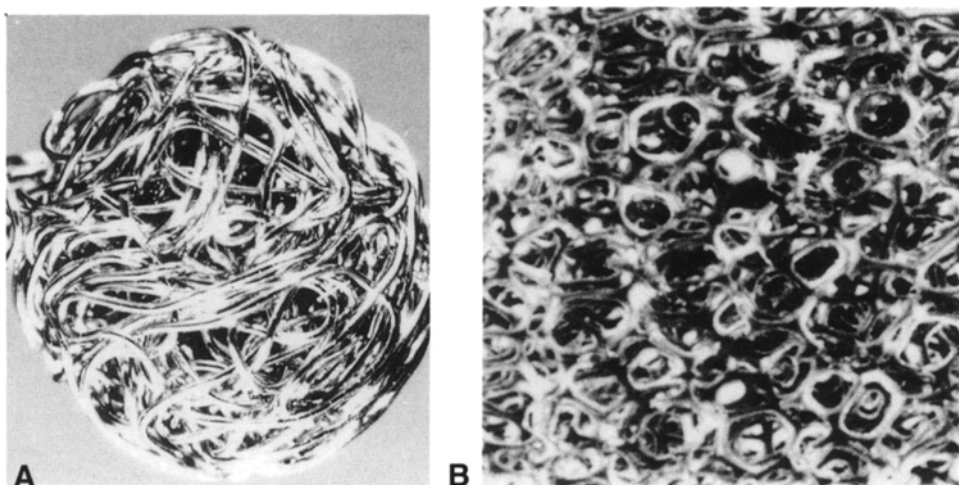


Fig. 1. Biomass support particles: (a) stainless steel wire mesh sphere; (b) reticulated foam cube.

voids within the particles are readily occupied by microorganisms forming films or flocs that subsequently become entrapped as a result of growth. Two major types of BSP are used, the reticulated foam cuboid and the stainless steel wire mesh sphere, *see* Fig. 1. These have been used with a range of microbial systems covering both growth associated (4) and non-growth associated products (5). Practical bioreactor systems developed for use with BSPs include the Fluidized Bed Reactor, the Spouted Bed Reactor, and the Circulating Bed Reactor (6). The Circulating Bed Reactor has been operated at scales of up to 160 m<sup>3</sup> for the treatment of industrial wastewater (the CAPTOR process, 7).

The biomass holdup in a particulate immobilized cell reactor may be defined as the product of the biomass holdup per particle ( $m_p$ ) and the number of such particles,  $N_p$ , per unit reactor volume,  $V_R$ . The volumetric reaction rate is thus given by

$$R_v = R_p m_p (N_p/V_R) \quad (1)$$

where  $R_p$  is the net specific rate of reaction for an immobilized cell particle.

The number of particles per unit reactor volume (the particle number density) will depend largely on physical parameters such as size, shape and density, hydrodynamic behavior, and mode of reactor operation. As such it is more a function of the choice of reactor than of the immobilization technique employed.

### The Circulating Bed Reactor

Many factors influence the choice of immobilized cell reactor type, principally the particle characteristics (size, shape, density, and robust-

ness). For particles that are essentially neutrally buoyant, for example, gel beads and foam BSPs, the Circulating Bed Reactor (CBR) of Black et al. (4) is particularly suitable. The CBR is a modification of a basic bubble column, and relies on airlift drive to bring about liquid and particle mixing. It was developed to exploit the advantages of cell immobilization using BSPs and is shown schematically in Fig. 2.

Two important parameters needed for the design of an aerobic immobilized cell reactor are the hydrodynamic behavior of the particles and the oxygen transfer performance. For stainless steel BSPs some information exists in the literature as to their fluidization behavior (8) (the fluidized bed being the favored reactor configuration for this type of particle). The mass transfer performance of similar systems has been studied by Ostergaard et al. (9,10) among others.

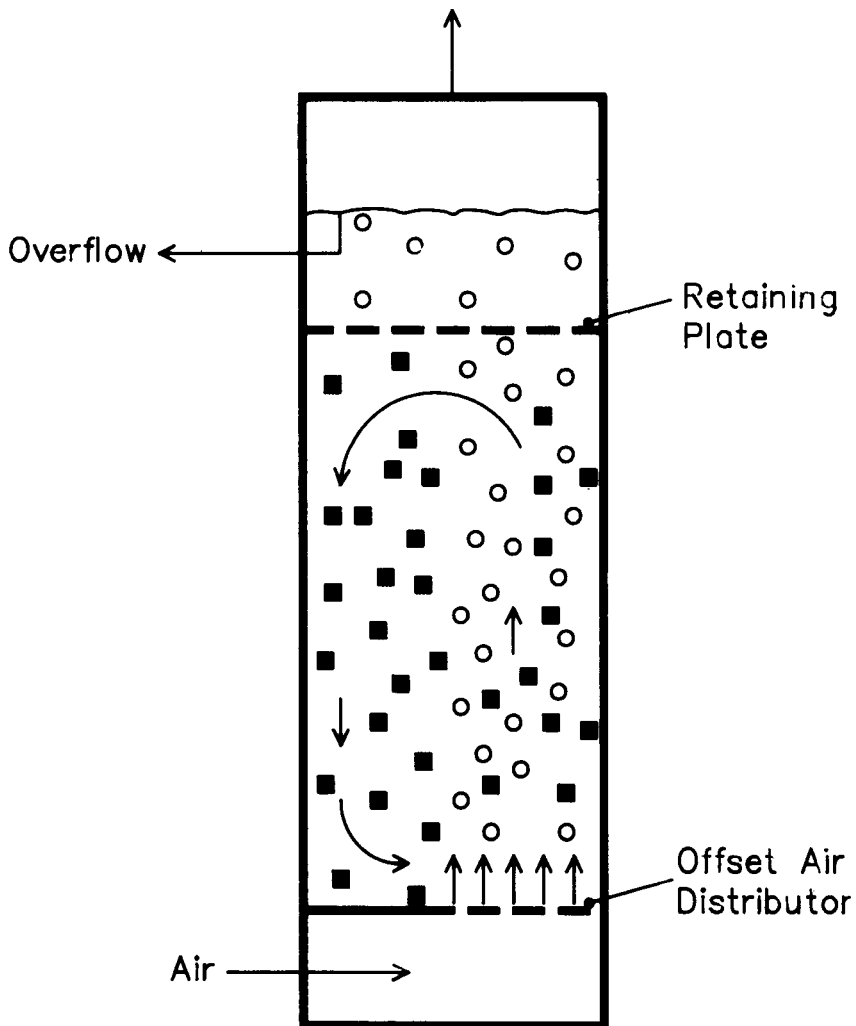


Fig. 2. The circulating bed reactor.

For the CBR, little information exists apart from the preliminary qualitative work of Teale et al. (11) concerning the effect of air distributor configuration and aspect ratio on particle circulation. For this reason, the present study was undertaken, being a study of the particle circulation and mass transfer performance of a CBR system using a range of particles of neutral buoyancy.

## PARTICLE CIRCULATION

It is often desirable when designing an immobilized cell reactor to maximize the particle number density so as to minimize the reactor volume. With many types of immobilized cell particles, particularly where cell growth occurs, it is necessary to operate in an expanded bed mode. With the CBR the bed expansion depends on the circulation of the particles. Factors affecting the particle circulation include superficial gas velocity, reactor configuration, size, shape, density, and surface condition of particle, gas distributor design.

Teale et al. (11) conducted a brief study of some of these parameters in a rectangular section tank. The particles used were CAPTOR type pads, filled with agar to simulate biomass. CAPTOR pads are cuboidal reticulated foam BSPs with dimensions  $25 \times 25 \times 10$  mm. The experiments, which were carried out with different distributor arrangements and aspect ratios, involved varying the number density of particles and measuring the minimum air flowrate required to achieve "good" mixing. It was found that, in general, irrespective of distributor design or aspect ratio, increased gas rates were required to circulate an increased particle number density. When total base aeration was used, the particles either remained at the bottom or top of the vessel. Distributor arrangements were therefore chosen to encourage circulation, and are shown in Fig. 3.

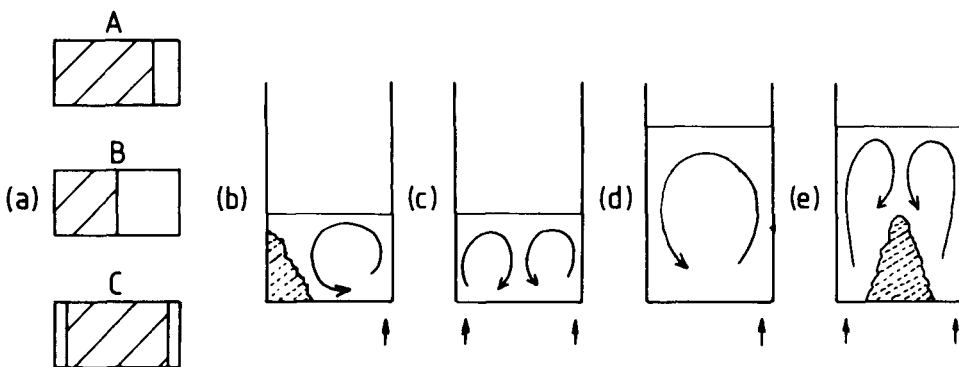


Fig. 3. Circulation in a CBR: (a) distributor configurations showing  $\phi = .2$ ,  $\phi = .5$ , and  $\phi = 2 \times .1$ ; (b) low aspect ratio, distributor A or B; (c) low aspect ratio, distributor C; (d) high aspect ratio, distributor A or B; (e) high aspect ratio, distributor C.

With the single sided aerators (A and B) it was found that the particle number density attainable in the tank increased with aspect ratio over the range studied. The opposite effect was observed, however, with the two sided aerators, i.e., lower aspect ratios allowed greater particle number densities.

It was suggested that circulation cells with a minimum aspect ratio of unity were best for particle circulation. Thus for low (less than 1) aspect ratios, two aerators were required, whereas one was sufficient for high aspect ratios. With single sided aerators, circulation was found to be best when the ratio of aeration area to base area ( $\phi$ ) was .5.

Although brief, this study showed that partial base aeration (i.e.,  $\phi < 1$ ) was superior to total base aeration, establishing circulation cells with a minimum aspect ratio of about one.

Beek (12) studied liquid vortices in similar gas-liquid circulating systems, and showed that the number of circulation cells would adjust so that the aspect ratio was approximately unity. Merry and Davidson (13) determined the optimum circulating aspect ratio in fluidized beds (gas-solid; liquid-solid) to be .8 although their particles were far denser than the BSPs used by Teale et al.

## Particle Circulation Studies

Particle circulation was studied in a tank with rectangular cross section of  $.30 \times .15$  m with  $\phi = .5$  at aspect ratios of up to 3.7, using a range of neutral buoyancy particles. The superficial gas velocity required to maintain circulation of all the particles was determined by reducing the gas velocity from a value at which all particles circulated well to one at which a stationary layer of particles began to form.

### *The Effect of Aspect Ratio*

Figure 4 shows nominal solids holdup plotted against minimum superficial gas velocity to circulate the particles for a range of aspect ratios. The particles used were polyethylene spheres (with a diameter of 15.5 mm and density of  $920 \text{ kg.m}^{-3}$ ). At aspect ratios less than unity, "dead zones" of static particles were observed in the upper corner of the tank opposite the air sparger. It was also observed that the section of the tank that remained circulating had an aspect ratio of about unity.

At aspect ratios of less than unity (where particles are required to move greater distances in the horizontal plane than in the vertical plane), full particle circulation was difficult to achieve. At the high gas velocities required to move the particles, much localized liquid turbulence was observed above the sparger area.

At higher aspects ( $> 1$ ), circulation was far easier to achieve. The particles had an essentially circular mixing pattern. The areas for upward and downward motion appeared to be equal, which is consistent with measurements of the liquid velocity in bubble columns (13). Increasing the aspect ratio above unity led to a slight reduction in the superficial gas velocity re-

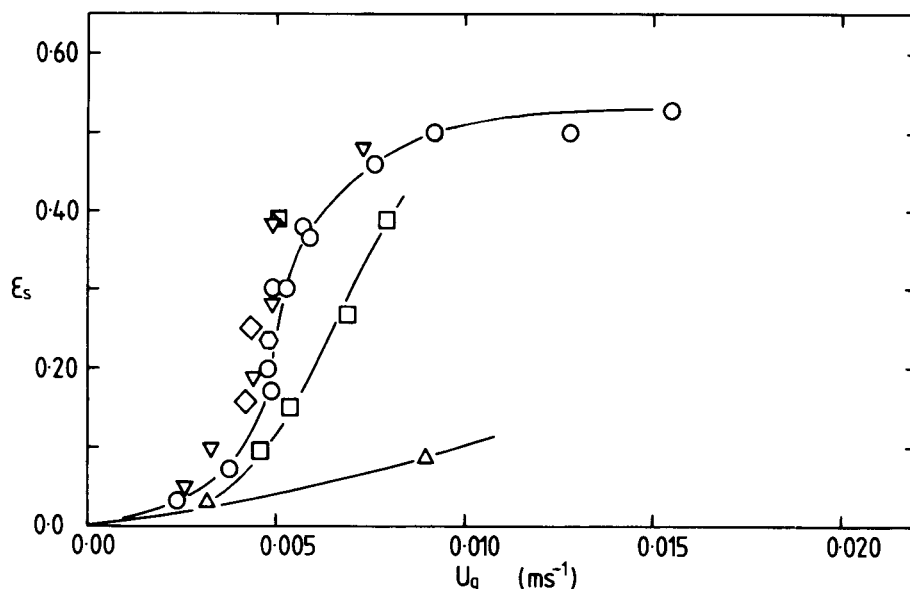


Fig. 4. Solids holdup,  $\epsilon_s$ , against superficial gas velocity required to circulate the particles,  $U_g$ , with a range of aspect ratios, for 15.5 mm polyethylene spheres. Aspect ratios: ( $\Delta$ ) .5; ( $\square$ ) .75; ( $\circ$ ) 1.0; ( $\nabla$ ) 1.5; ( $\diamond$ ) 2.0; ( $\diamond$ ) 3.0; ( $\boxtimes$ ) 3.7.

quired to move a given solids holdup. This, too, is consistent with the observation (14) that the liquid circulation velocity in the riser of an airlift contactor increases with liquid dispersion height.

#### *The Effect of Particle Properties*

Figure 5 shows the apparent solids holdup plotted against minimum superficial gas velocity required to circulate the particles, at an aspect ratio of unity. A range of neutral buoyancy particles were used including BSPs filled with agar to simulate biomass. Some similar data is reported in the literature for operating Circulating Bed Reactors. This is included for comparison.

All the spherical particles behaved similarly, but the 13.0 mm particles were slightly easier to circulate than the 15.5 and 18.4 mm particles. The maximum solids holdup attainable was in the region of .55. These spherical particles have a packed bed voidage ( $1 - \epsilon_s$ ), of approximately .45, thus the maximum particle holdup possible would be approximately .55, as indicated in Fig. 5. It would appear that it is in order to achieve circulation it is necessary to expand the bed by only a small degree.

Far lower superficial gas velocities were required to circulate both the empty and agar filled BSPs. The literature data (4, 7), which was obtained using CAPTOR type particles containing activated sludge, and 6 mm cubic BSPs containing yeast agree well with the agar filled BSP data. The yeast filled BSP data was obtained in a CBR system with an aspect ratio of 3.4, but as can be seen in Fig. 4, aspect ratio has little effect above unity. The

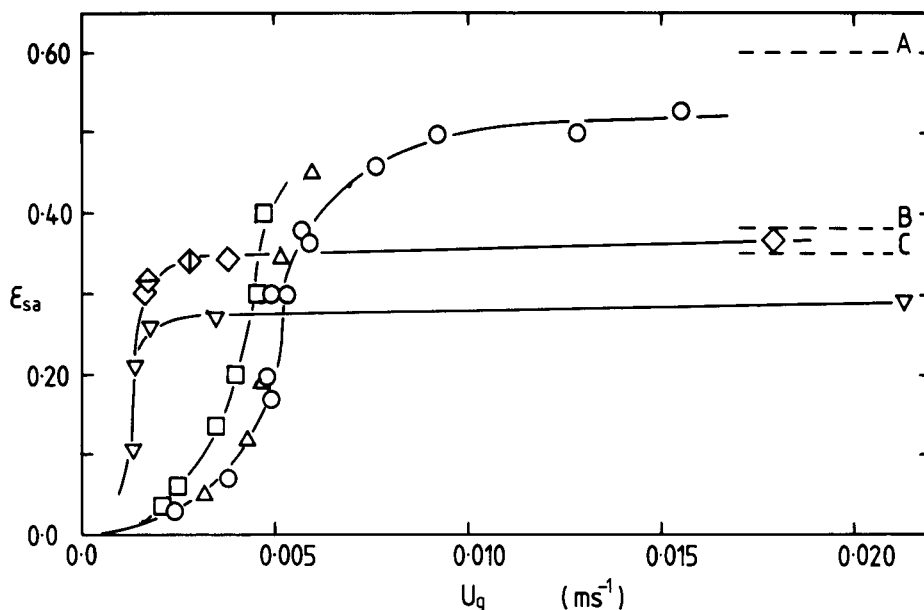


Fig. 5. Apparent solids holdup,  $\epsilon_{sa}$ , against superficial gas velocity required to circulate the particles,  $U_g$ , at an aspect ratio of unity with a range of particle types: ( $\square$ ) 13.0 mm polyethylene spheres; ( $\circ$ ) 15.5 mm polyethylene spheres; ( $\triangle$ ) 18.4 mm polyethylene spheres; ( $\nabla$ ) 10 mm cubic BSPs, empty; ( $\diamond$ ) 10 mm cubic BSPs, agar-filled; ( $\diamond$ ) from Ref. 4; ( $\diamond$ ) from Ref. 7. Lines A, B, and C represent packed bed solids holdup for polyethylene spheres, agar-filled BSPs and empty BSPs, respectively.

difference in the superficial gas velocity required to move the various particles is possibly caused by their differing terminal settling/rising velocities,  $U_t$ . These were found experimentally and are given in the following table, along with other particle properties. As the terminal settling/rise velocity of the BSPs is far lower than those of the spherical particles, then less energy (from the sparged gas) could be expected to be required to achieve circulation.

The maximum solids holdup of BSPs achieved was less than that of the spherical particles. Slightly higher values than those obtained with the

Table 1  
Particle Properties

Particle	Size, mm	Density, $\text{kg.m}^{-3}$	$U_t$ , $\text{m.s}^{-1}$
Polyethylene spheres	18.4	924	.174
Polyethylene spheres	15.5	920	.157
Polyethylene spheres	13.0	930	.137
Agar filled cubic BSPs	10	1020	.036
Empty cubic BSPs	10	1002	.022



empty BSPs are attainable with the agar filled BSPs. The packed bed voidage for these particles was found to be approximately .65 and .62, respectively, indicating a maximum solids holdup of .35 and .38, respectively (see Fig. 5). The maximum circulating solids holdup can be seen to approach the packed bed solids holdup, but again a degree of bed expansion is required to allow circulation.

## OXYGEN MASS TRANSFER

The oxygen mass transfer rate,  $N_R$ , to a pneumatically sparged contactor is given by Eq. [2].

$$N_R = k_L a (C_L^* - C_L) \quad (2)$$

If the gas phase is present as bubbles with a Sauter mean diameter of  $d_b$ , then the specific interfacial area may be estimated from

$$a = 6\epsilon_g/d_b \quad (3)$$

where  $\epsilon_g$  = mean volumetric gas holdup.

Combining the above two relationships allows identification of the main parameters governing gas-liquid mass transfer

$$N_R = k_L (6\epsilon_g/d_b) (C_L^* - C_L) \quad (4)$$

In a bubble column the mean bubble size ( $d_b$ ) depends partly on the initial size of bubbles generated at the sparger, and partly on the coalescent properties of the liquid. Low viscosity bubble columns can, however, be reasonably divided into two distinct groups; coarse bubble systems where the mean bubble size is between four and six mm, and fine bubble systems where the mean bubble size is much less than six mm (15). In general, fine bubble systems only result from the use of noncoalescing media and then only when used with fine bubble spargers such as porous diffusers or two phase injectors. The aeration plate used in the present study was designed to produce coarse bubbles following the recommendations of Fair (16) and consisted of seven .5 mm holes drilled through 3 mm perspex sheet.

For values of  $d_b$  less than approximately 2 mm the bubble surface can be rigid or mobile, depending on the surface-active materials in the liquid. This can lead to variations in the liquid film mass transfer coefficient,  $k_L$ . However, above 2 mm the bubble surface is always mobile and  $k_L$  is essentially independent of surface active materials. For coarse bubbles in air water systems,  $k_L$  is usually between  $3 \times 10^{-4}$  and  $4 \times 10^{-4}$  m.s<sup>-1</sup>.

The mean gas holdup,  $\epsilon_g$ , is defined as the volume fraction of gas in the two or three phase mixture. It is a function mainly of the superficial gas velocity and is sensitive to the liquid properties.

At low superficial gas velocities, with uniform aeration of the base, the bubbling is termed quiescent and is characterized by a reasonably uni-

form distribution of evenly sized bubbles. For the case of a batch liquid bubble column, the gas holdup is given by

$$\epsilon_g = U_g/U_{bs} \quad (5)$$

The single bubble rise velocity  $U_{bs}$ , is dependent upon bubble size and water purity, but in the range 3–8 mm is reasonably constant at  $.235 \text{ m.s}^{-1}$ . As the superficial gas velocity is increased, channeling occurs with larger bubbles formed by coalescence passing up the center of the column. This results in radial holdup profiles and internal circulation (18).

Studies including the effects of large (greater than 1 mm) particulate solids on the mass transfer performance of aeration equipment reported in the literature have been confined largely to dense solids in three phase fluidized beds, where the particles were considerably denser than the fluidizing medium (eg glass ballotini, density  $2600 \text{ kg.m}^{-3}$ ) (18–21). Very little work has been carried out with particles of low relative density. Recently, however, Verlaan et al. (22) has shown that the presence of alginate beads and polystyrene spheres (2.5 mm diam) had little effect on the mass transfer performance of an external loop airlift. Walker et al. (7), however, reported that oxygen mass transfer was improved by the presence of CAPTOR particles in a circulating bed system.

## Oxygen Mass Transfer Studies

Two features of the Circulating Bed Reactor (CBR) system that distinguish it from a bubble column, and which could be expected to affect its gas-liquid mass transfer characteristics are the induced uniform circulation of liquid, and the presence of particles.

These were studied separately in various configurations of a rectangular section (.15 × .075 m) system at a fixed aspect ratio of 2.4 and the results compared with a uniformly aerated bubble column in an attempt to better understand the behavior of the CBR.

Oxygen transfer rate was determined using the sulphite oxidation technique of Cooper et al. (23). The volumetric mass transfer coefficient,  $k_{La}$ , was evaluated using the method of Danckwerts (24) to calculate a value of  $C_L^*$ . More extensive details of the experimental apparatus and procedure for oxygen transfer rate determination are given in (22).

### Basis of Results

The oxygen transfer rate, as measured by a technique relying on liquid analysis such as sulphite oxidation, produces a transfer rate based on the liquid volume,  $N_L$ , with the units  $\text{kmol (O}_2 \text{ transferred) .m}^{-3} \text{ (liquid) .s}^{-1}$ . In order to assess utilization of reactor volume a more useful basis is  $N_R$ , which has the units  $\text{kmol (O}_2 \text{ transferred) .m}^{-3} \text{ (reactor) .s}^{-1}$ . These two transfer rates are related by

$$N_R = N_L (1 - \epsilon_s - \epsilon_g) \quad (6)$$

where for porous particles  $\epsilon_s = \epsilon_{sa} (1 - \psi)$ ,  $\epsilon_{sa}$  is the apparent solids holdup,  $\psi$  is the particle porosity ( $\text{m}^3 \text{ void} \cdot \text{m}^{-3} \text{ particle}$ ), and

$$\epsilon_{sa} = N_P V_P / V_R \quad (7)$$

where  $V_P$  = enclosed particle volume.

### The Particle Free Bubble Column

Figure 6 shows results for oxygen transfer rate,  $N_R$ , plotted against superficial gas velocity,  $U_g$ . The solid line is that given by linear regression analysis of the data. In the range considered,  $N_R$  is seen to be a linear function of the superficial gas velocity. The shaded area is the range of oxygen transfer rate estimated by Eqs. [4] and [5] using typical values of  $U_{bs}$ ,  $k_L$ , and  $d_b$  given in (15). The data is within this shaded area indicating that  $k_L$  was constant and that the specific interfacial area was a linear function of the superficial gas velocity.

### Particle Free Circulating System

Figure 7 shows the oxygen transfer performance of a particle free circulating system. As superficial gas velocity was increased, the oxygen transfer rate increased linearly initially, showing a very similar relationship to that in Fig. 6. At about  $6.7 \times 10^{-3} \text{ m} \cdot \text{s}^{-1}$  however, this relationship changed abruptly. The transfer rate remained linear with superficial gas

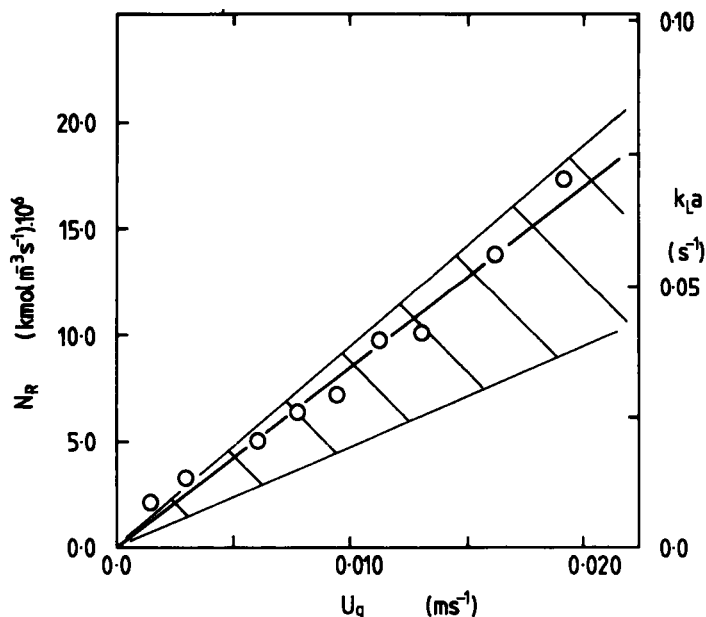


Fig. 6. Oxygen transfer rate based on reactor volume,  $N_R$ , and mass transfer coefficient,  $k_L a$ , against superficial gas velocity,  $U_g$ , in a particle free bubble column. Shaded area represents typical range of values from the literature.

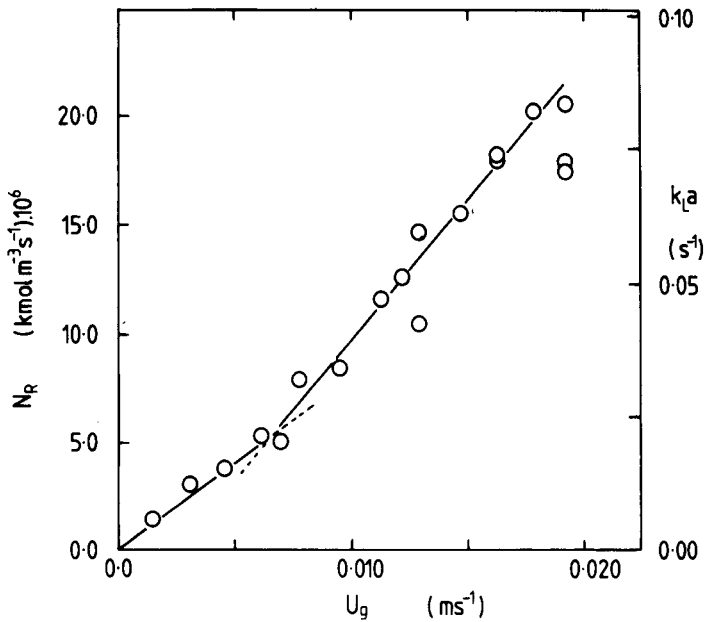


Fig. 7. Oxygen transfer rate based on reactor volume,  $N_R$ , and mass transfer coefficient,  $k_{La}$ , against superficial gas velocity,  $U_g$ , in a particle free circulating system.

velocity, but the gradient of the line became steeper. This may be explained in terms of visual observations made during the experiments. At lower values of  $U_g$ , (below  $5 \times 10^{-3} \text{ m.s}^{-1}$ ) liquid circulation could not be discerned and the bubbling was very similar to that obtained with full base aeration (bubble column). The only difference between this system and the bubble column at this stage was that the bubbles were closer together, because of the arrangement of the sparger orifices. As superficial gas velocity was increased, the liquid began to circulate because of air lift drive (at approximately  $5.8 \times 10^{-3} \text{ m.s}^{-1}$ ) as evidenced by the downward movement of some of the smaller bubbles in the non-aerated half of the column. Entrainment of this sort begins when the downward velocity of the liquid equals the terminal rise velocity of the smallest bubbles. Freedman and Davidson (1969) observed the entrainment of 1 mm bubbles into the downflow of a concentric cylinder airlift when bubbles of 3–8 mm were being generated at the sparger. They also observed that when using a bubble column of similar dimensions with partial base aeration, the gas holdup was comparable to that obtained with full base aeration at superficial gas velocities of up to  $20 \times 10^{-3} \text{ m.s}^{-1}$ . The increase in oxygen transfer beyond this point in Fig. 7 was therefore probably a result of the entrained bubbles that had the effect of lowering the mean bubble diameter and increasing the specific interfacial area.

At higher circulation rates more bubbles were entrained into the downflow and a certain degree of bubble classification could be seen. Since

bubbles could leave the downflow and reenter the riser section at any point along the column, or coalesce and rise against the liquid flow, only the smallest bubbles were drawn right down to the bottom of the downflow.

### *Circulating Bed of Particles*

The oxygen transfer characteristics of a circulating bed of solids, with a nominal solids holdup of .30 was investigated. The nominal solids holdup was based on the enclosed volume of the particles (*see* Eq. [8]) and as such represented a dimensionless particle number density.

The oxygen transfer rate based on reactor volume,  $N_R$ , is shown against superficial gas velocity in Fig. 8. The solid line shown is that obtained by linear regression analysis. It can be seen that over the ranges considered, particle size and particle shape had similar decreasing effects on the oxygen transfer rate (approx 30%, compared with the bubble column). Entrainment of gas bubbles into the downflow was observed, as in the particle-free system, although the mean bubble size appeared greater. The rise of the bubbles against the liquid flow was additionally hindered by the downward movement of a bed of particles. In the case of BSPs, they tended to entrap gas bubbles, which then remained within the particles.

### *Effect of Solids Holdup on Oxygen Mass Transfer*

A series of experiments was performed to investigate the effect of solids holdup on oxygen transfer rate in a circulating bed. The superficial gas velocity was held constant at  $9.3 \times 10^{-3} \text{ m.s}^{-1}$ , while the solids holdup

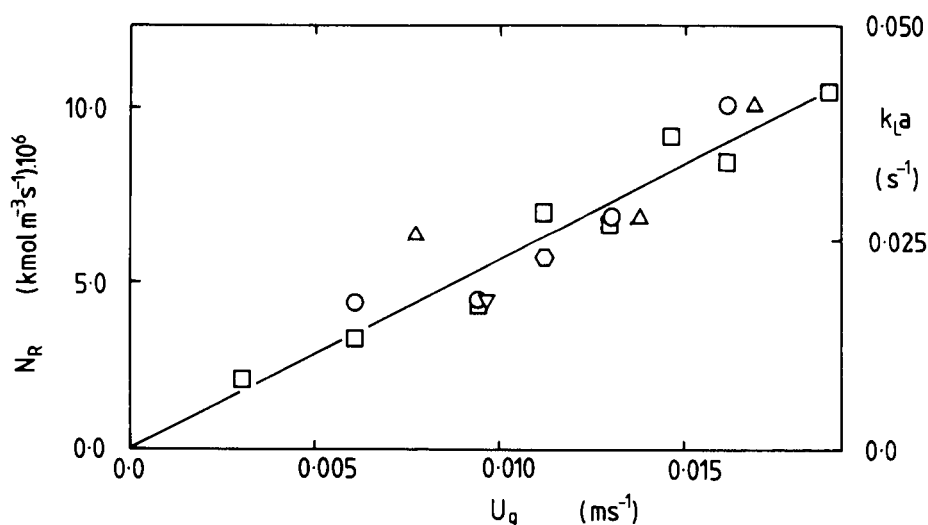


Fig. 8. Oxygen transfer rate based on reactor volume,  $N_R$ , and mass transfer coefficient,  $k_La$ , against superficial gas velocity,  $U_g$ , for a circulating bed of particles at an apparent solids holdup of .3 with a range of particle types: ( $\square$ ) 6 mm empty cubic BSPs; ( $\circ$ ) 13.0 mm polyethylene spheres, ( $\circ$ ) 15.5 mm polyethylene spheres; ( $\triangle$ ) 18.4 mm polyethylene spheres; ( $\nabla$ ) 4.5 mm calcium alginate gel beads.

was varied from zero to .60 (using 13 mm spheres) and zero to .30 (using 6 mm BSPs).

The oxygen transfer rate based on reactor volume  $N_R$  is shown in Fig. 9 against apparent solids holdup,  $\epsilon_{sa}$ . It can be seen that at low solids holdup values (up to .10) the presence of the spherical particles led to an increase in oxygen transfer rate. The behavior of the tank contents appeared very similar to that in the solids free system, with about the same degree of bubble entrainment in the downflow. The spherical particles did not appear to cause the bubbles to coalesce. As the solids holdup was increased the oxygen transfer rate began to fall until it reached a minimum at a solids/particle holdup of approximately .30-.45, at which point the particles appeared to have a definite bubble coalescing effect. The actual transition from noncoalescing to coalescing was not, however, observed. Above a solids holdup of approximately .45, particles began to pack at the top of the tank. At this stage the oxygen transfer rate was observed to increase again. This was probably because of an increased gas holdup in the packed cap and an increased tendency for bubbles to recirculate.

The presence of BSPs had the effect of gradually decreasing oxygen transfer rate to a similar value as the spheres at a holdup of .30-.35, at which point bubble coalescence was again observed.

## DISCUSSION

It can be seen that the two important design parameters, namely, the circulating particle number density and the oxygen transfer rate, are both

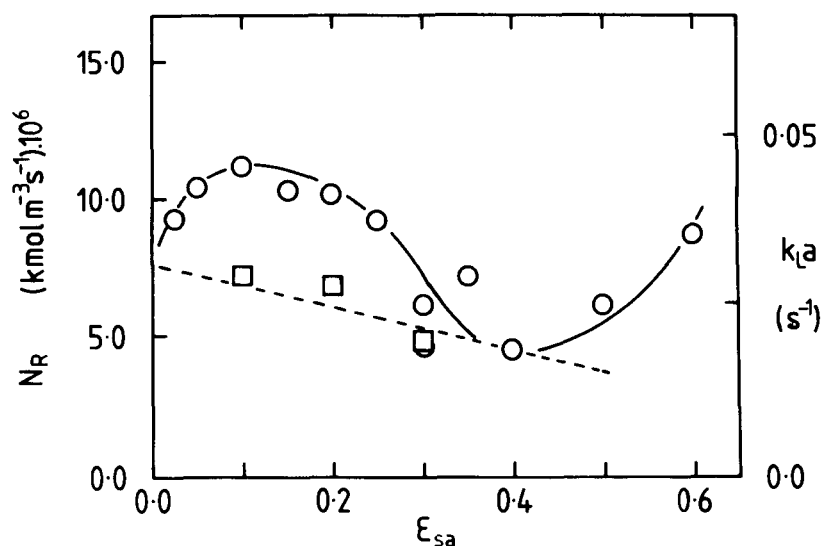


Fig. 9. Oxygen transfer rate based on reactor volume,  $N_R$ , and mass transfer coefficient,  $k_{La}$ , against apparent solids holdup,  $\epsilon_{sa}$ , at a superficial gas velocity of  $9.3 \times 10^{-3}$  for (○) 13.0 mm polyethylene spheres and (□) 6 mm empty cubic BSPs.

dependent upon the superficial gas velocity. Whether the superficial gas velocity required to circulate a given number of particles is also sufficient to maintain the desired metabolism of the immobilized organism, will depend on the actual biological system. Figure 10 shows the oxygen transfer rate per unit of solid volume plotted against solids holdup. When presented in this form, the oxygen transfer rate represents the amount of oxygen available to the immobilized cell particles. The rate of oxygen demand of such particles depends on

1. The maximum specific microbial oxygen uptake rate. Some values can be found in the literature, though these are usually for free cell systems, and may well be different for immobilized cells.
2. Diffusional gradients within the particles of both oxygen and substrate. This can be accounted for by the use of effectiveness factors.

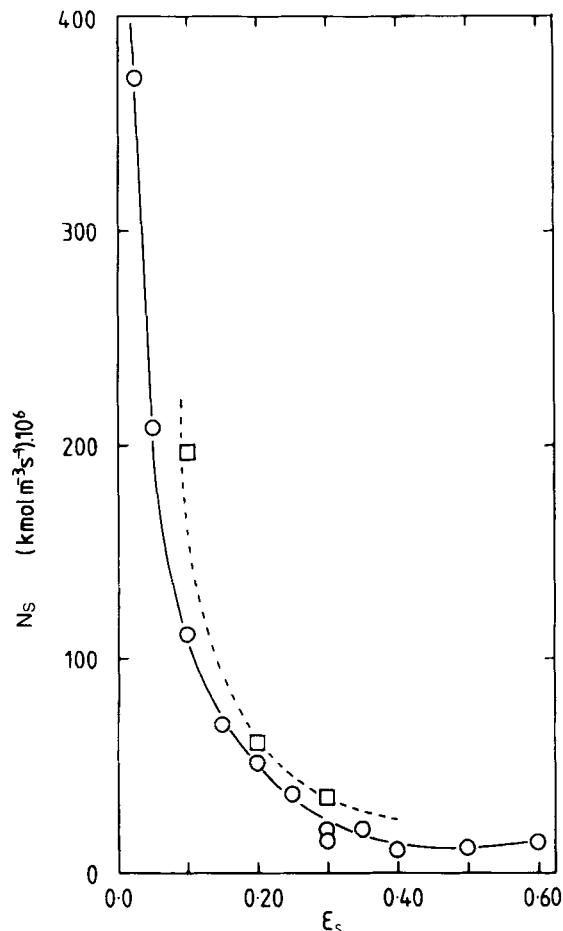


Fig. 10. Oxygen transfer rate based on solids volume,  $N_s$ , against solids holdup,  $\epsilon_s$ , for a circulating bed of 13.0 mm polyethylene spheres at superficial gas velocities of (○)  $9.3 \times 10^{-3} \text{ m.s}^{-1}$  and (□)  $16.2 \times 10^{-3} \text{ m.s}^{-1}$ .

3. The biomass holdup per particle. This will be affected by such biological parameters as cell packing density, and may be affected by operational parameters such as hydrodynamic shear. It is ultimately limited by the potential volume fraction of the particle that can be occupied by biomass. This is equal to .97 for foam BSPs and .30 for calcium alginate gel beads (24).

An actively respiring free cell yeast culture has an oxygen consumption rate of around  $80 \times 10^{-6} \text{ kg(O}_2\text{) .kg}^{-1} \text{ (dry wt) .s}^{-1}$ , and yeast has a packed cell density of approximately  $250 \text{ kg (dry wt) .m}^{-3}$  (wet cells). If it is assumed that particles contain such yeast cells and that no diffusional limitations exist, then for maximum biomass holdup the consumption rate would be about  $650 \times 10^{-6} \text{ kmol (O}_2\text{) .m}^{-3} \text{ (particle) .s}^{-1}$  for BSPs and  $200 \times 10^{-6} \text{ kmol (O}_2\text{) .m}^{-3} \text{ (particle) .s}^{-1}$  for gel beads. With reference to Fig. 10, it can be seen that only a very small number of particles could be supplied with oxygen, whereas Fig. 5 shows that a far larger number of particles could be circulated in the reactor. This, however, represents "the worst possible case," since it is unlikely that all cells immobilized in such a way would be respiring at similar rates to freely suspended cells.

The above example identifies two superficial gas velocities. One is the velocity required to circulate a given number of particles (*see* Fig. 5), and the other is the velocity required to supply sufficient oxygen to the particles. If the latter is greater than the former, then the chosen mode of reactor operation could be to operate at the superficial gas velocity required to circulate the particles, but increase the oxygen transfer rate (e.g., by using pure oxygen or external aeration); or to operate at a superficial gas velocity above that required to circulate the particles, in order to supply sufficient oxygen.

The final choice will depend on the particular system requirements and the overall cost.

## CONCLUSIONS

The superficial gas velocity required to circulate a given number of particles would appear to be related to the terminal settling/rise velocity of the particles, which is a function of particle size, shape, density, and surface condition. The maximum particle holdup is limited by the packed bed voidage, and for BSPs this depends of whether the particles are empty or filled.

Oxygen mass transfer in a particle free circulating system was better than in a bubble column at superficial gas velocities sufficient to cause bubble entrainment into the downflow. The effect of neutral buoyancy particles on the oxygen transfer characteristics of such a circulating system varied with solids holdup and particle porosity. A low solids holdup of nonporous particles led to enhanced oxygen transfer rates, whereas the presence of porous particles led to reduced rates.



It is unlikely that, for highly aerobic microbial systems, the CBR could supply sufficient oxygen to the organism at the superficial gas velocity required for particle circulation. At the high superficial gas velocities necessary to provide sufficient oxygenation, problems may be encountered with maintaining the integrity of the immobilized cell particles. For anaerobic systems, e.g., ethanol production by yeast and those with low oxygen demand (plant cells), the CBR is, however, a practical form of immobilized cell bioreactor.

## ACKNOWLEDGMENT

The authors are grateful for financial support provided by the SERC.

## REFERENCES

1. Bucke, C. (1986), "Techniques for Cell Immobilization," In Webb, C., Black, G. M., and Atkinson, B., (eds.), *Process Engineering Aspects of Immobilized Cell Systems*, Rugby, I. Chem. E. Publ., pp. 20-34.
2. Fonseca, M. M. R. da, Black, G. M., and Webb, C. (1986), "Reactor Configurations for Immobilised Cells," In Webb, C., Black, G. M., and Atkinson, B., (eds.), *Processing Engineering Aspects of Immobilized Cell Systems*, Rugby, I. Chem. E. Publ., pp. 63-74.
3. Atkinson, B., Black, G. M., Lewis, P. J. S., and Pinches, A. (1979), "Biological Particles of Given Size, Shape, and Density for Use in Biological Reactors," *Biotech. Bioeng.* **XXI**, pp. 193-200.
4. Black, G. M., Webb, C., Mathews, T. M., and Atkinson, B. (1984), "Practical Reactor Systems for Yeast Cell Immobilization Using Biomass Support Particles," *Biotech, Bioeng.* **XXVI**, 134-141.
5. Fukuda, H., Webb, C., and Atkinson, B. (1984), "Continuous Cellulase Production in a Spouted Bed Fermenter Using Cells Immobilized in Biomass Support Particles," *Third European Congress on Biotechnology*, vol. 1, Weinheim, Verlag-Chemie, pp. 547-552.
6. Black, G. M., and Webb, C. (1984), "A Practical Technology for Cell Immobilization based on Novel Biomass Support Particles," *Proc. VI Australian Biotechnology Conference*, Uni. Queensland, Brisbane Sept. 5-7.
7. Cooper, P. F., Walker, I., Crabtree, H. E., and Aldred, R. P. (1986), "Evaluation of the CAPTOR Process for Updating an Overloaded Sewage Works," In Webb, C., Black, G. M., and Atkinson, B., (eds.), *Process Engineering Aspects of Immobilized Cell Systems*, Rugby, I. Chem. E. Publ., pp. 205-217.
8. Webb, C., Black, G. M., and Atkinson, B. (1983), "Liquid Fluidization of Highly Porous Particles," *Chem. Eng. Res. Des.* **61**, pp. 125-134.
9. Ostergaard, K., and Suchozebrski, W. (1971), "Gas-liquid mass transfer in gas-liquid fluidized beds," *Proc. 4th Europe. Symp. Chem. Reaction Eng.*, Pergamon, Oxford, 1971, p. 21.
10. Ostergaard, K., and Fosbol, P. (1972). "Transfer of Oxygen across the Gas-Liquid Interface in Gas-Liquid Fluidized Beds," *Chem. Eng. J.* **3**, pp. 105-111.

11. Teale, M. J., Hemsley, P., and Smith, K. (1981), "Some Observations on the Hydrodynamics of Foam Biomass Support Particles," undergraduate project report, Dept. of Chem. Eng., UMIST, Manchester, UK.
12. Beek, W. J. (1965), "Oscillations and Vortices in a Batch of Liquid Sustained by a Gas Flow," *Symposium on Two-Phase Flow*, Univ. Exeter, UK.
13. Merry, J. M. D., and Davidson, J. F. (1973), "Gulf Stream Circulation in Shallow Fluidized Beds," *Trans. I. Chem. E.* **51**, pp. 361-368.
14. Joshi, J. B., and Sharma, M. M. (1979), "A Circulation Cell Model for Bubble Columns," *Trans. I. Chem. E.* **57**, pp. 244-251.
15. Onken, V., and Weiland, P. (1981), In Moo-Young, M., Robinson, C. W. and Vezina, C., (eds.), *Advances in Biotechnology*, vol. 1. *Scientific and Engineering Principles*, Pergamon, Toronto, p. 559.
16. Heijnen, J. J., and van't Riet, K. (1984), "Mass Transfer, Mixing and Heat Transfer Phenomena in Low Viscosity Bubble Column Reactors," *The Chem. Eng. J.* **28**, B21-B42,
17. Fair, J. R. (1973), In Perry, R. H., and Chilton, C. H., (eds.), *Chemical Engineers Handbook*, McGraw-Hill Kogakusha, Tokyo, pp. 18-72.
18. Hills, J. H. (1974), "Radial Non-Uniformity of Velocity and Voidage in a Bubble Column," *Trans. I. Chem. E.* **52**, pp. 1-9.
19. Verlaan, P., Holst, A. C., Tramper, J., van't Riet, K., and Luyben, K. Ch. A.M. (1984), *Third European Congress on Biotechnology*, vol. 1, Weinheim, Verlag-Chemie, p. 151.
20. Cooper, C. M., Fernstrom, G. A., and Miller, S. A. (1944), "Performance of Agitated Gas-Liquid Contactors," *Ind. Eng. Chem.*, **36**, **6**, pp. 504-509.
21. Dankwerts, P. V. (1970), *Gas-Liquid Reactions*, McGraw-Hill, London, p. 19.
22. Dean, J. F. (1985), *Oxygen Transfer Studies in Gas Agitated Systems*, M. Sc. thesis, Victoria University of Manchester, UK.
23. Freedman, W., and Davidson, J. F. (1969), "Hold-up and Liquid Circulation in Bubble Columns," *Trans. I. Chem. E.* **47**, **8**, pp. 251-263.
24. Webb, C. (1986), "Biomass Hold-up in Immobilized Cell Reactors," In Webb, C., Black, G. M., and Atkinson, B., (eds.), *Process Engineering Aspects of Immobilised Cell Systems*, *I. Chem. E. Publ.* pp. 117-133.