OXYGEN TRANSFER IN A THREE-PHASE FLUIDISED BED BIOREACTOR WITH NOVEL BIOMASS SUPPORT WHEN USED IN INDUSTRIAL WASTEWATER TREATMENT

PART II: EFFECT OF SUPPORT MASS AND LIQUID HEIGHT

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Abstract

The effect of support mass and liquid height on the oxygen transfer in the three-phase fluidised bed were experimentally investigated. Novel biomass support, the KMT^R, with density of 850 kg/m^3 and an equivalent size, d_p = 8.3 mm, has been used in this work. The oxygen transfer rate was expressed in terms of the volumetric oxygen transfer coefficient, k_La. It has been found out that the $_Lk$ a values increased with increasing support mass up to a critical solids load of 4.0 kg, above which the k_La decreased and that k_La decreased with increasing liquid height. It has been recommended that for a given bed height, the three-phase fluidised bed bioreactor should be operated at an experimentally determined critical solid mass, so as to increase the oxygen transfer rate. A good agreement between experimental and literature data is revealed in this paper.

Introduction

Cocurrent three-phase fluidised beds, where the solid particles are fluidised by both the liquid (continuous phase) and gas (dispersed phase) are used in processes which require good contacting between solid, liquid and gaseous phases. Examples of applications are catalytic hydrocracking, gas absorption and heterogeneous catalytic hydrogenation [1], in biological fermentation [2], and recently, in biological wastewater treatment [3]. Biological treatment of wastewater is by far the most commonly used form of treatment. While in activated sludge systems bacteria grows in suspension in aerated tanks, in the three-phase fluidised bed bioreactors, a mixed culture of micro-organisms' growth takes place on the surface of fluidised or suspended particles called biomass support.

Recently, advances in both the basic understanding of the biotechnology and the hardware technology have enabled to overcome many of the problems associated with the anaerobic processes. Among the newer technological advances have been the biological fluidised bed (BFB) processes [4], where wastewater and compressed air are passed upward through a bed of media at velocities sufficient to reduce the bulk density of the liquid so that light particles floating on the liquid surface can sink and distribute equally throughout the liquid. The biological treatment of industrial wastewater using the BFB process is a newly developed concept. Relying on the typical biological organisms found in other treatment works, it owes its high rate success to much higher surface area and biological mass (biomass) concentration than those in conventional processes. The BFB can be used for aerobic carbonaceous BOD removal and nitrification, anaerobic carbonaceous BOD removal and anoxic denitrification of industrial and municipal wastewater[5].

Advantages of the fluidised beds over fixed beds are obvious, not only in the wastewater treatment practice, but also in catalytic reactions. In the fluidised state, the biomass support are relatively easy to handle, a property which favours continuous flow and recirculation systems. Fluidised beds are mechanically simple and suited to large-scale operations. Fluidised beds tend to be more compact than alternative phase contacting techniques and offers high heat and mass transfer surfaces with high respective transport coefficients under comparable flow conditions.

The principal disadvantages of the three-phase fluidised bed system used in this work include the following: the flow of liquid and gas was concurrent, leading to unfavorable effect on the driving force for mass transfer. To approach a countercurrent flow, a multi-compartment reactor was required which could lead to more expenses than fixed beds. With the low height to diameter ratios there may result appreciable longitudinal mixing of phases in the reactor resulting into low conversion rates. Scale-up from laboratory- to commercial-sized reactors is not simple because different regimes of fluidisation tend to occur at different scales of operation, that is, small diameter beds tend to slug, moderate-sized beds are freely bubbling and large-diameter beds exhibit large scale circulation. For any size of BFB, these regimes must be predicted and allowed for.

Economic studies made by Wheeldon and Bayley[6] indicated that for carbonaceous and nitrogenous oxidation, BFB system is competitive with the activated sludge process [7]. In their study, they examined a number of process configurations over a range of different flow rates. They costed the following processes in an oxygenic BFB for sewage treatment: (a) carbonaceous oxidation, (b) carbonaceous oxidation and nitrification and, (c) carbonaceous oxidation, nitrification and denitrification in a two-BFB systems. For each case they compared the cost of the BFB process with that for building and operating a conventional activated sludge process to produce the same effluent quality, using a net present value technique to cover both capital and operating costs. However much of the data used were derived from a laboratory bioreactor operated at steady flow rates. Therefore further research is needed to investigate large scale bioreactors and to provide the data to refine the economic analysis.

The basis of the three-phase fluidised bed process is the very large surface area per unit volume (specific surface) which is available for growth of very high concentration of micro-organisms [8]. Thus, support loading is a key factor for the specific surface. Due to conflicting reports from different workers on the effect of support mass on oxygen transfer, it was intended in this work to study the effect of support mass and establish the critical support loading, M_{sc}, for the KMT^R support.

The volumetric oxygen transfer coefficient, k_La, depends among other factors on the gas-phase hold-up, ϵ_g, which in turn depends on the aeration rate. Presence of biomass support in the fluidised bed reduces the gas-phase hold-up and the flow area of the gas and liquid phases, leading to increased bubble coalescence and in a reduced oxygen transfer rate. The way k_La depends on the amount of support charged into the bioreactor has been reported to depend on equivalent size of the support, d_p, [3]. Joly *et al.*[8] report that solids loading in the three-phase fluidised bed affects the k_La in the same way as the catalyst loading affects the overall mass transfer rate in the catalytic hydrogenation [8].

Presence of the biomass support in the bioreactor controls the bubble size, bubble rising velocity and the spiral liquid circulation rate caused by the air lift action [9]. A change in rheological properties of water and wastewater due to presence of solids has also been reported by Jadhav and Pangarkar [10], as per equation

$$\mu_{rel}=\mu_{mix}/u_L=1+2.5\varepsilon_s+10\varepsilon_s^2+0.0027\exp(16.6\alpha) \quad (1)$$

where the subscripts: *rel, mix, L* and *s* stands for relative, mixture, liquid and solid, respectively, and α is the alpha-factor, defined by Foster and Wase [11], as follows:

$$\alpha=(k_fa)_{wastewater}/(k_fa)_{cleanwater} \quad (2)$$

Since k_La is inversely proportional to the liquid viscosity, increasing ϵ will increase the viscosity and thereby lower k_La. It was the objective of this research to find out experimentally how the KMT^R support loading affects the k_La. The amount of biomass support charged into the three-phase fluidised bed, size and density are, however not the only important parameters for designing a bioreactor.

Variations in particle shape, entrained air, porosity, surface configuration, etc., are other factors which together make it impossible to design a standard system to suit all requirements.

On the other hand, the performance efficiency of any biological reactor containing support particles depends upon the number of support particles per unit reactor volume, the average biomass hold-up per particle, the average overall specific rates of reaction of biomass and on the overall yield coefficients of the immobilized biomass. To achieve higher oxygen transfer rate, a pre-selected quantity of support, and hence of biomass could be accumulated in the bioreactor leading to a rational basis for design as compared with trickling filters and biodiscs. Use of the biomass support in wastewater treatment systems is of paramount importance due to the following advantages: biomass is retained within the aeration tank, hindering the need for recycle; very high biomass concentrations can be maintained compared to dispersed growth systems, thus smaller reactor volumes are required for the carbonaceous oxidation stage; sludge can be removed directly from the support reducing the clarifier loading [12].

Despite of the physical difference between three-phase fluidised beds and two-phase and slurry bubble columns, similar observations concerning the effect of solid particles on k_La have been reported in these reactors as shown in Table 1. The effect of the support on k_La has also been reported to depend on the nature of the biomass support used [1], necessitating the need for establishing the relation for any type of support under use.

While testing the effect of solids loading on k_La in olefinic polymerization process conditions, Li *et al*[13], found out that the k_La values for the three gases (propylene, ethylene and hydrogen) in liquid n-hexane increased at low solid concentration (10 wt%) and decreased at high solid concentration (30 wt%). The literature survey on the effect of solids on k_La is presented in Table 1 for air in different liquids using different types of solids at ambient temperature and pressure. From Table 1, it can be seen that the effect of solids load on k_La is dependent of nature of both liquid and solids under test.

The interaction between complex hydrodynamics and transfer mechanism complicates analysis, design and prediction of the three-phase fluidised bed systems. This is because, firstly, at present, there is no systematic approach to the quantitative description of transport mechanism, and secondly, prediction of transport processes under gas induced three-phase fluidisation is still poor, as compared to gas fluidised bed behavior which is reported in several works and reviews [17].

Table 1

Reference	Liquid	Solid	Remark
Mehta and Sharma[14]	Na_2CO_3(aq), DEA	$CaCO_3$, $BaCO_3$	k_La decreased at lower solids loading and increased at higher solids loading
Oguz *et al.*[15]	Water	Soda glass ballotini (at 3 - 20 wt%)	k_La decreased at 20 wt%
Chapman *et al.*,[16]	Water	Sea sand (<80 μm); Kieselguhr (<50	k_La decreased with increasing solid loading

Winkler[18], expressed k_La in terms of the operating parameters of the bioreactor, where it can be seen that k_La is proportional to the liquid height, H_L, in some expressions, while other equations do not show dependency of k_La on H_L [18]. In this work, the k_La values have been measured experimentally for a range of liquid depths, to establish the exact relationship for the designed pilot plant utilizing the KMT^R support in industrial wastewater treatment. Winkler expressed the oxygen transfer rate at different liquid levels by considering the fraction of oxygen molecules which diffuses into the liquid from the air bubble, as the later rises from the bottom of the column to the top. He defined the fraction as the oxygen utilization efficiency, E_o. He reported that E_o increases with the liquid height due to increased bubble residence time, as per equa

$$E_0=(H_{L1}/H_{L2})^{0.7} \tag{3}$$

where E_o is given by

$$E_0=100(1-n_s/n_0)\% \tag{4}$$

In equation (4), n_o and n_s are the numbers of moles of oxygen in the bubble initially and on arrival at the liquid surface. In the aeration of a detergent solution with an alpha-factor similar to sewage mixed liquor, for several designs of aerators, the oxygen transfer rate per unit tank area (that is kg oxygen/m^2.h), increased with liquid depths between 1 and 8 m [18]. This shows that the higher the liquid height the higher the gas residence time and the higher the oxygen transfer. Hence H_L is another design factor for oxygen transfer rate, and selection of the aspect ratio for the bioreactor (L/D), must put into consideration the residence time of the air bubbles.

The effect of liquid height on air hold-ups have been studied also by Tsuchiya and Nakanishi [19], while varying the number of holes on the distributor plate. With multi-orifice plate (as in this work), a non-negligible difference between the two heights has been reported. It was found out that the scheme of correlation covering the distinctive bubble regimes, i.e. the dispersed, clustered or coalesced and the slugging bubble regimes favours liquid at greater depths.

While studying the effect of level on gas hold-ups, Kochbeck *et al.*[20], found out that the highest local gas hold-up in a three-phase reactor occurs at the top of the reactor regardless of the solid loading and the gas input. The increase of the local gas hold-up along the axis was attributed to the decreasing hydrostatic pressure. Further studies revealed that solids loading in the range 1.4 to 5.1% by volume indicated only a negligible influence on the local gas hold-up.

The objective of the study presented in this paper was to simultaneously study the effect of support mass and liquid height on oxygen transfer in the three-phase fluidised bed bioreactor when used in industrial wastewater treatment. The research was carried out through the following steps: designing and assembling of the bioreactor; designing and construction of the air and liquid distributors; selection of a biomass support for wastewater treatment; selection and assembling of the bioreactor accessories; and assembling of the experimental rig. Experiments were conducted after testing and commissioning of the rig.

Experimentation

Experimental Set-up

Figure 1 shows the schematic diagram of the pilot plant. The bed was made by introducing a known weight of support into the column, through branch (8). Liquid from the feed tank was pumped into the column to a required height of slurry, H_{SL}. For fluidised column heights below V_7, liquid recirculation was not possible; for H_c maintained at V_7 or above, the valve V_7 was set in such a way that the liquid flow past the valve counterbalanced the pumping rate to maintain the column height. While keeping the liquid flow rate constant, compressed air was introduced to fluidise the bed, and varied stepwise, from u_g = 3.7 mm/s, just below u_{mf} = 8 mms^{-1}, up to a maximum of 42.5 mms^{-1}. The bed was aerated for 3 to 5 minutes before data was collected. Oxygen concentration was measured by using dissolved oxygen probes (DOP), inserted direct into the column (4).

Operational range: The bioreactor was operated in the range of 3.7 to 42.5 mm/s for air velocity; while the liquid velocity ranged from 0 to 6.6 mm/s. Compressed air pressure was varied between 2 to 5 bar, while the liquid height was varied between 1.0 to 4.0 m. Solids loading was varied from 0.5 to 5.5 kg. The total fluidised bed height, H_c, varied between 1.0 to 3.5 m.

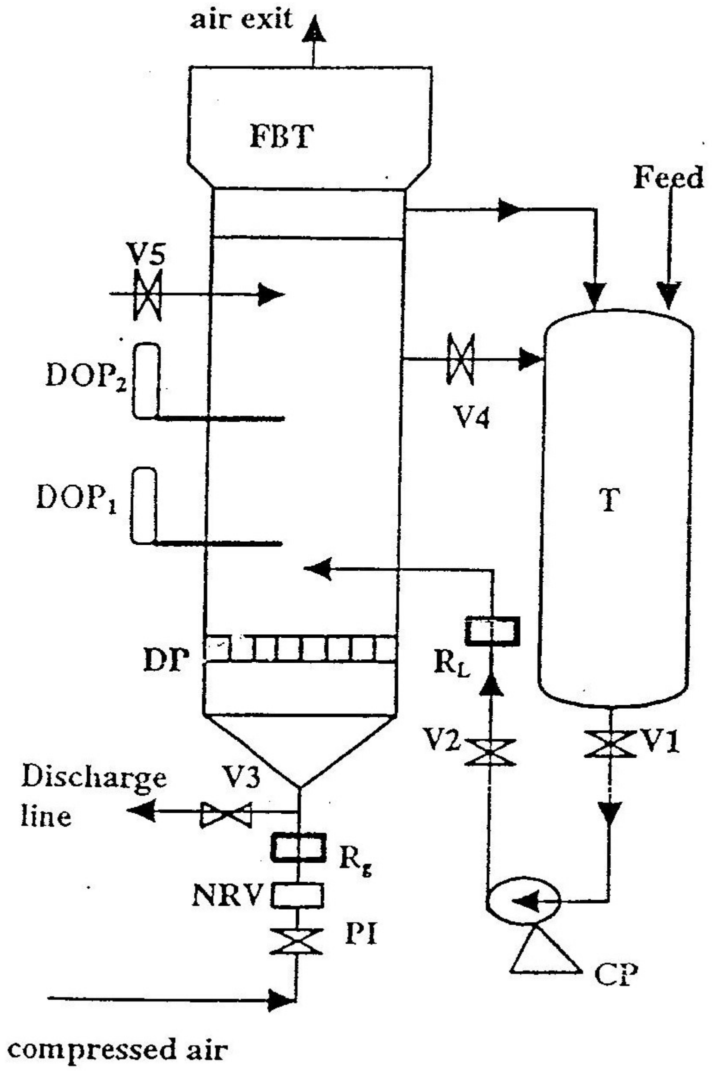


Fig. 1 Schematic of the pilot plant

3.2 Methodology

The bed expansion, E_b, was determined from the bed height difference before and after aeration, as given in the equation

$$E_b = \Delta H / H_{sL} \tag{5}$$

The air hold-up, ϵ_g, is the fraction of the bed volume occupied by air, determined from the equation

$$\varepsilon_g = \Delta H / H_c \tag{6}$$

The $(k_La)_h$ was determined from the hydrodynamic parameters of the bioreactor as per the following equations:

$$(k_{La})_h = 1.154(P_0/V_R) \tag{7}$$

where

$$P_0/V_R = g[A - B + C]/\varepsilon_L \tag{8}$$

and $A = \rho_s \epsilon_s (u_L + u_g)$; $B = \rho_L u_L (1 - \epsilon_L)$; $C = \rho_L u_g \epsilon_L$ [21].

The liquid- and solid-phase hold-ups were determined as per equation

$$\varepsilon_L = M_L/(H_{cA_c}\rho_l) \tag{9}$$

and

$$\varepsilon_s = 1 - \varepsilon_g - \varepsilon_L \tag{10}$$

The $(k_La)_{CT}$ was determined from direct measurements of oxygen concentration, and applying the CSTR model, which employs an equation

$$(k_La)_{CT} = u_L(C_2 - C_1)/[\Delta Z(C_g^* - C_2)] \tag{11}$$

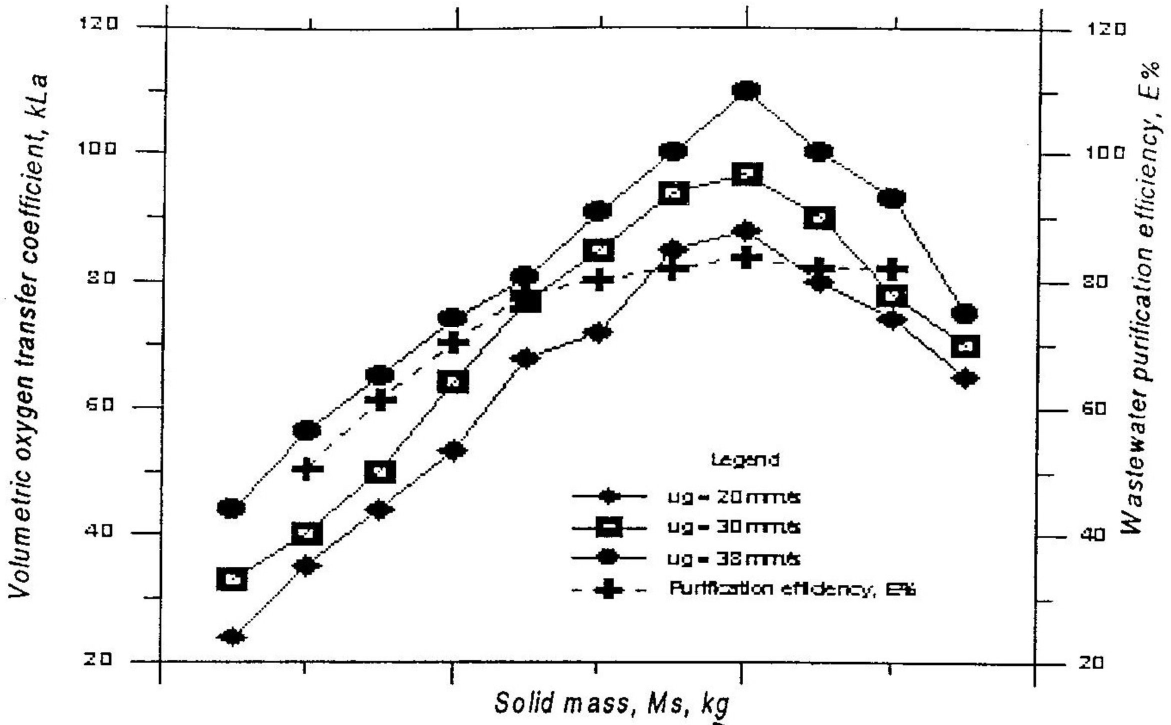
Discussion

In this section, results and observations are presented. Literature findings have been cited for comparison and further reading.

4.1 Effect of Solid Mass

It was observed that as M_s was increased from 0.5 kg to 4.5 kg, the k_La was initially increasing, as shown in Fig. 2(a), (up to 4 kg for a liquid height of 2.0 m), beyond which a decrease in k_La was observed. This trend was observed at all air flow rates, as shown in Fig. 2(b). The increase in $(k_La)_h$ with increasing mass of support is supported by similar results obtained in this work, that wastewater purification efficiency increased initially with increasing mass of support, up to 4.0 kg, at bed height of 2.0 m. Beyond a solid loading of 4.5 kg, a poor phase mixing at a constant height and a decrease in k_La caused by accumulation of support at the top of the bed, was observed. From Fig. 2 it can be said that the k_La was strongly affected by support mass at all air flow rates.

The above findings were attributed to the fact that increasing solid loading in a



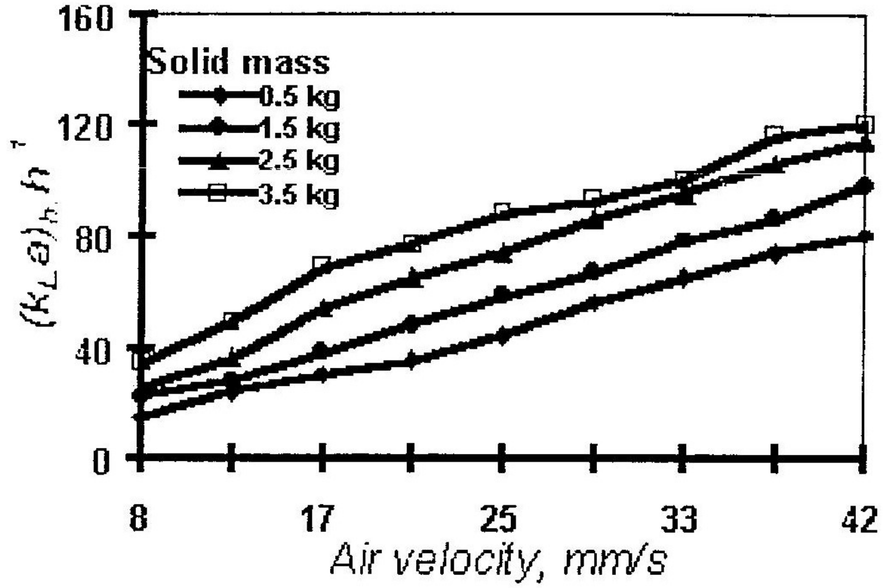


Fig. 2 (a) Variation of the volumetric oxyegen transfer coefficient (k_La)h with solids loading, M_s, at fixed bed height of 2.0 m, and liquid velocity, u_L=4mm/s. Effect of Ms on waste water purification efficiency, E, is also depicetd on a plot. (b) effect of solid mass on k_La-u_g relationship

three-phase fluidised bed at a fixed bed height beyond M_{sc} lowers gas hold-up, ϵ_g, which is an index for the mean residence time of the air in the bioreactor. High solid loads lowers air hold-up due to a hindered bed expansion. The increase in k_La at low concentration of solids was attributed to the phenomena governing the interaction between gas bubble and solid particles. That is, if the Weber number (the ratio between the inertial force to the interfacial force) is higher than 3.0, then the particle is able to divide or stretch the bubble and the interfacial area increases. Also with the larger size of the KMTR support, there is a greater chance for the particle to create turbulence effect on the gas-liquid film, whose thickness decreases leading to high oxygen penetration rate [18]. At higher solid loading, the kinetic energy of the air was not sufficient to mix the phases homogeneously, and low k_La values were obtained. Similar results have been reported by Lu *et al.*[21], and Karamanev *et al*[22]. Detailed study of the k_La-M_s relationship revealed that at each liquid height there was a maximum support mass that gave highest k_La, called the critical biomass loading, M_{sc}, as shown in Fig. 3(a). The critical biomass support loading was again found to decrease with increasing liquid height as shown in Fig. 3(b), that is, there was a shift in peak on the k_La-M_s curves from right to left as H_L was increased from 1.5 to 4.5 m.

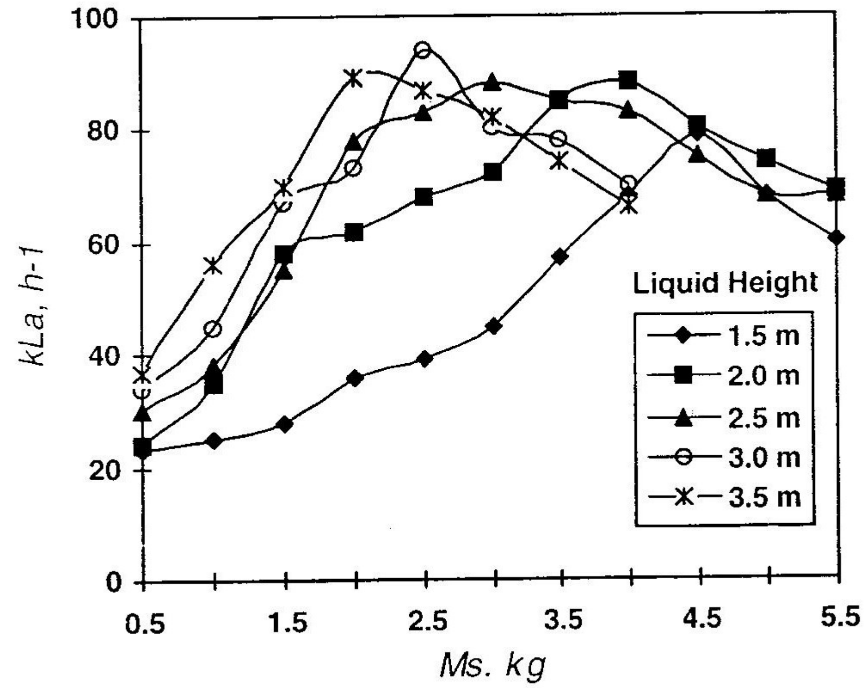


Fig. 3 (a) Effect of support mass on volumetric oxygen transfer coefficient, k_La: Shift in critical support loading with increasing liquid height, H_L

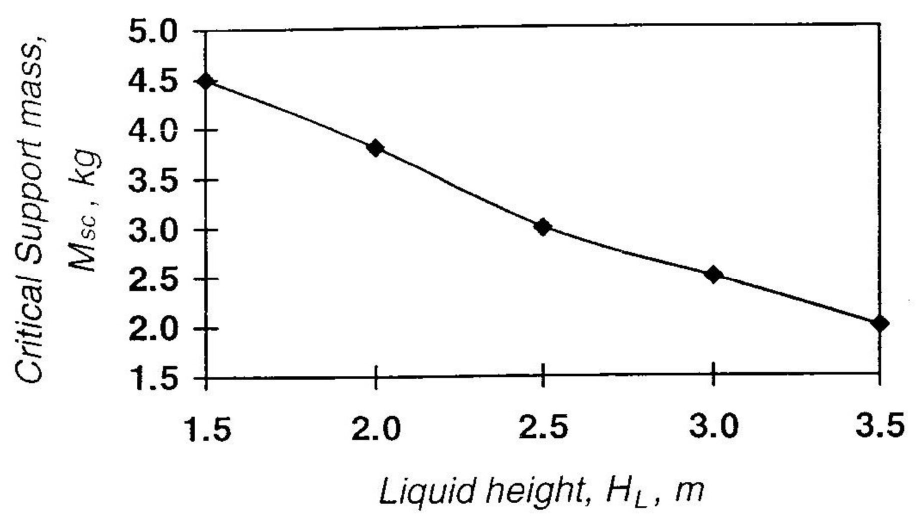


Fig. 3 (b) Effect of support mass on volumetric oxygen transfer coefficient, k_La, Critical biomass support masses at different liquid heights.

Improvement of k_La in the bioreactor is therefore possible by operating the bed with solids loading equal to M_{sc}, which has to be determined experimentally for a given type of biomass support. If conditions necessitate use of solids loading beyond M_{sc}, then improvement of oxygen transfer rate in a fluidised bed with a limited bed height can be achieved by using air lift-loop bioreactor connected sideways, acting as downcomer, which improves air hold-up and turbulence in the bed, as suggested by Kochbeck *et al.*[20].

The critical solid loading, M_{sc}, was found to be 4.0 kg (precisely, within the range from 3.5 to 4.5 kg), at a constant liquid height of 2.0 m, as shown in Fig. 2(a), while Fig. 2(b) shows how the M_{sc} values varied with liquid heights. Results presented in Fig. 2 agrees with literature reported by Joly *et al.*[8] and Li *et al.*[13], that an increase in k_La was observed at solids loading lower than 5 - 10% by weight, and that beyond this critical value, k_La decreased considerably.

Increasing the amount of solids charged into the bioreactor resulted into an increase in solid-phase hold-up, ϵ_s, if the bioreactor height remains constant. A plot of k_La versus ϵ_s, Fig. 4(a), shows an increase in k_La as ϵ_s increases, while Fig. 4(b) shows that increasing the liquid phase hold-up lowers k_La.

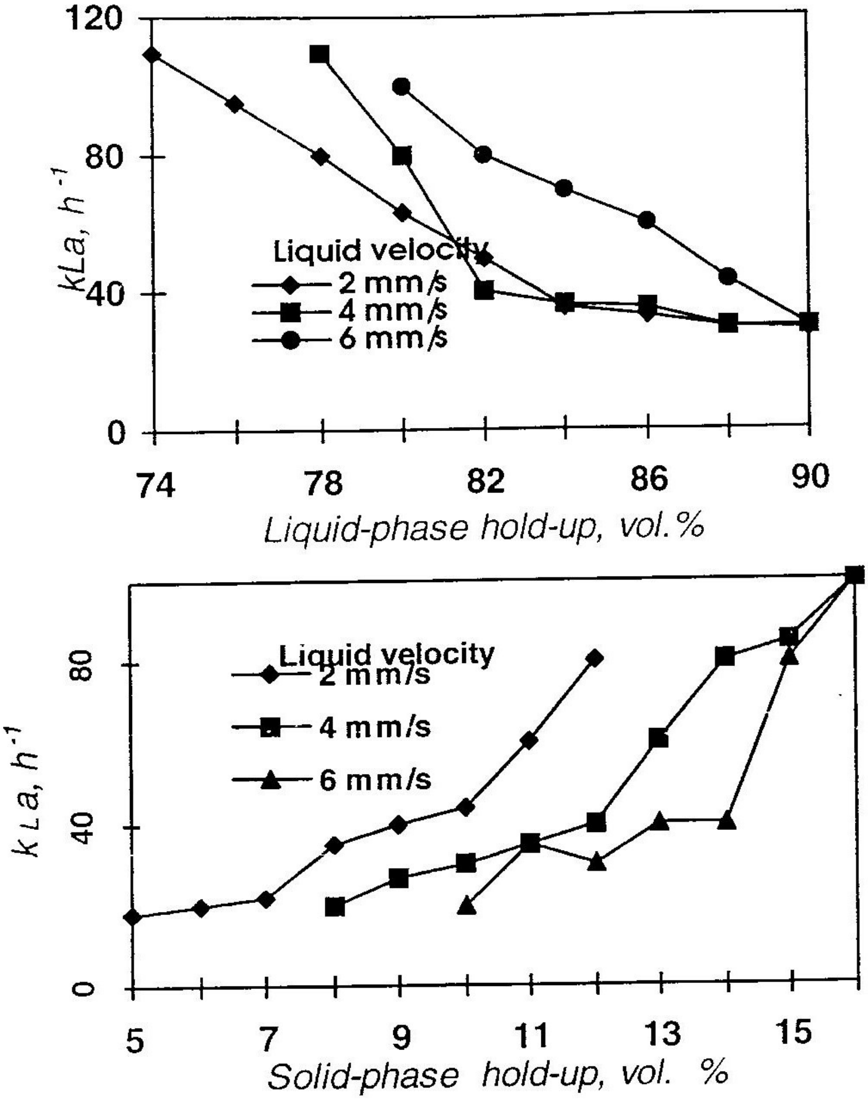


Fig. 4 Variation of k_La with the liquid- and solid-phase hold-ups

4.2 Effect of Liquid Height

It was observed in this work that k_La was decreasing as the liquid height was increased from 1.00 to 4.50 m. The results were attributed to poor scouring effect of the air at higher liquid heights. To avoid the decrease in k_La it has been suggested by Fan *et al.*[23], to stage two or more three-phase fluidised beds with only one air inlet at the bottom of the lower unit, where he found out that for three-phase bioreactors, k_La values were higher in the two stage than in the single stage. Figure 5 gives the values of $(k_La)_h$ at various air velocities and liquid heights. As can be seen from Fig. 5, the $(k_La)_h$ values decreased as the liquid height was increasing. Increasing the liquid height from 1.0 m to 2.6 m, decreased the $(k_La)_h$ by 50%. It can also be seen from Fig. 5 that H_L affects $(k_La)_h$ strongly. The results have been attributed to the fact that increasing H_L increases the hydrostatic head on the gas bubble emerging from the distributor hole, reducing its kinetic energy and its agitating effect.

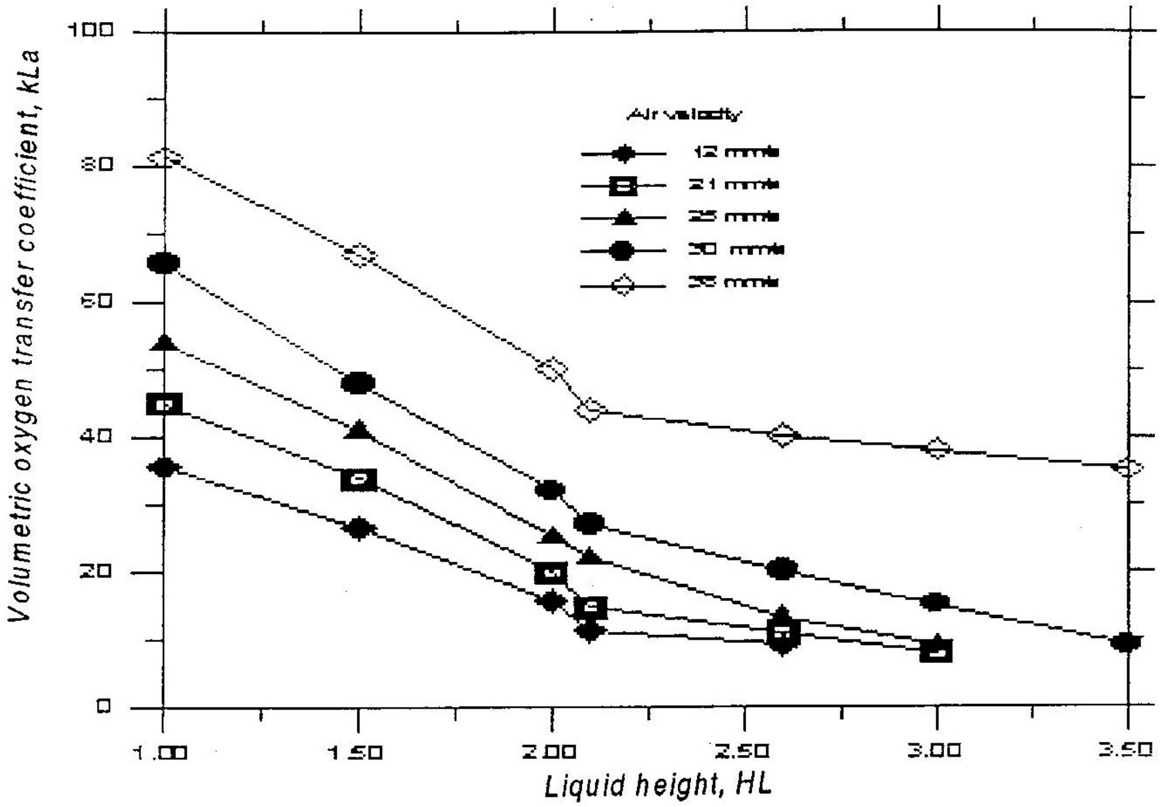


Fig. 5 Effect of liquid height, H_L, on volumetric oxygen transfer coefficient, k_La, at various air flow rates and at a fixed mass of support, M_s = 2.0 kg

The advantages of using relatively large diameter particles in a three-phase fluidised bed is that the small gas bubbles produced give a large interfacial area for mass transfer. Figure 6 shows measurements of volumetric oxygen transfer coefficient, $(k_La)_{CT}$ from equation (11), for a bed of KMT[R] biomass support, at different levels of the bed, Z, when compared with literature values [21],[24]. The literature data reported by Lee and Buckley[20], Ostergaard and Fosbol[23], are higher than the experimental values due to different liquids being used. While industrial wastewater was used in determining experimental data, Lee and Buckley used octanol solution.

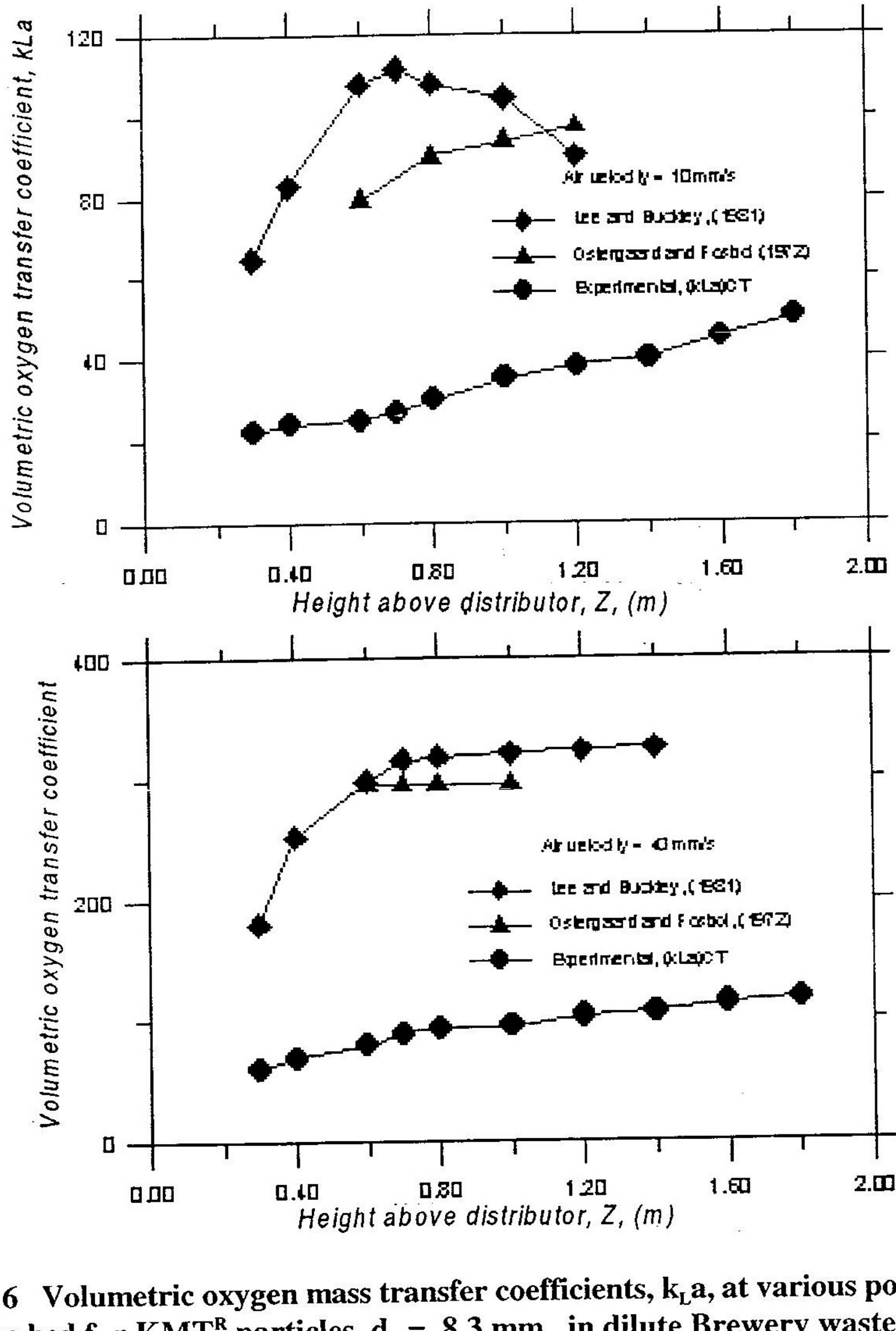


Fig. 6 Volumetric oxygen mass transfer coefficients, k_La, at various positions in the bed for KMT[R] particles, d_p = 8.3 mm, in dilute Brewery wastewater, at u_L = 4 mm/s. Literature data is included for comparison.

The design of the gas distributor was such that the gas was introduced as fairly large bubbles which were broken up by the particles. Thus in Fig. 6, the lower values of k_La at the base of the bed are mainly due to somewhat larger bubble size and smaller interfacial area in this region. Measurements of DOC at different levels above the distributor plate were achieved by dipping the oxygen probe (DOP2) into the column to a desired level, giving the value of C_2 while C_1 was measured at a fixed position, 10 cm above the distributor plate.

It has been also reported by Chen *et al.*[25], that the air hold-up increases with the axial position of along the bed. The increase in air hold-up and k_La along the bed height is attributed to the fact that: (a) bubbles get broken as they rise from the distributor, reducing in size and increasing the interfacial area, (b) schemes of flow regime vary with axial distance from the distributor plate as established by Chen *et al.*[25].

5 CONCLUSIONS

From the above findings, it can be concluded that:

For each design of the three-phase fluidised bed utilizing the KMT^R support, the critical biomass support, M_{sc}, has to be determined and applied in normal operation.

Since H_L lowers k_La, it is concluded that design of bioreactors should aim at using shallow wide beds, instead of tall narrow columns, although a compromise between residence time and low k_La effect must be satisfied.

With the above conflicting effects of M_s and H_L on k_La, a compromise must be reached from experimentation, so as to obtain the optimum combination of two operating parameters. Since the biomass concentration depends on the amount of solids charged and the two depends on the amount of liquid in the bioreactor, the importance of obtaining the optimal combination becomes obvious.

The data obtained in this work compares well with literature findings and can be used for design of industrial bioreactors.

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NOMENCLATURE

A_c - cross-sectional area of the column, m^2.
C - dissolved oxygen concentration, mg/l
C_g* - saturation dissolved oxygen concentration, mg/l
dp - equivalent size of the biomass-free support particle, mm
E_0 - oxygen utilization efficiency from the gas bubble
g - acceleration due to gravity, m/s_2
H_c - column height at fluidised state, m
H_L - gas-free liquid height in the column, m
H_{sL} - gas-free slurry height (liquid and solid), m
k_La - volumetric oxygen mass transfer coefficient, h^{-1}
$(k_La)_h$ - k_La from hydrodynamic parameters
$(k_La)_{CT}$ - k_La from oxygen concentration measurements
M_s - solid mass, kg
M_{sc} - critical solid mass, kg
M_L - liquid mass, kg
n - number of moles of oxygen in the gas bubble
P_0 - power consumption in aeration, W
V_R - volume of the bioreactor, m^3
Z - axial distance from the distributor, m

Greek letters
α - alpha-factor
ϵ - phase hold-up
ρ - density, kg/m^3

Subscripts
s, L, g - solid, liquid and gas, respectively