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Oxygen Mass Transfer in Bubble Column Bioreactor

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RESEARCH ARTICLE

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Abstract

In the present study volumetric oxygen mass transfer coefficient k_La has been determined for biodegradation of phenol in a bubble column bioreactor. Experimental studies have been carried out at different i) feed concentrations of phenol, ii) air flow rates and iii) feed flow rates. Dynamic method has been used to determine the oxygen mass transfer coefficient. The mass transfer coefficient for oxygen obtained is in the range of 0.00513 – 0.01793 s^{-1}. k_La was found to increase with increase in air flow rates and decrease with increase in feed concentration of phenol. The values obtained in this work are compared with the values available in literature. A mathematical correlation is derived for k_La in terms of dimensionless numbers.

Keywords

Biodegradation · bioreactor · bubble column · mass transfer · oxygen · phenol

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1 Introduction

Oxygen transfer mass transfer from air to the broth is very important in the design and operation of bioreactors degrading phenol. Aerobic organisms need oxygen for growth, cell maintenance and product formation. Different methods for measurement of k_La are available in literature [1]: sodium sulfite oxidation method, dynamic gassing-out technique, direct measurement and dynamic method. The dynamic method has been the most widely used measurement technique for bioreactors. The method is popular due to its simplicity and accuracy [2].

Bubble column bioreactors have a number of advantages in terms of in design and operation as compared to other reactors. They have excellent heat and mass transfer characteristics. Little maintenance and low operating costs are required due to lack of moving parts. Bubble columns have been investigated for gas holdup [3,4], bubble characteristics [5,6], flow regime investigations and computational fluid dynamics studies [7,8], local and average heat transfer measurements, [9,10] and mass transfer studies [11].

The majority of oxygen transfer data for airlift / bubble column bioreactors have been obtained with air/water-based systems. It is known that both liquid viscosity and surface tension affect volumetric oxygen transfer coefficient (k_La) airlift bioreactors containing water-based media [12]. Though many studies have been reported using bubble columns in bioprocesses, there have been no reports of biodegradation of phenol in bubble column bioreactor. In this study biodegradation of phenol has been carried out in a bubble column bioreactor. In bubble column bioreactor oxygen is required for the biodegradation of phenol. It is supplied in the form of air. Oxygen from gas phase dissolves in the liquid phase, where biodegradation reaction takes place. To model the oxygen transfer from gas phase to the bulk phase the knowledge of volumetric oxygen transfer coefficient k_La is required. Table 1 gives some of the correlations from literature for the volumetric coefficient for different reactor systems. It can be seen that different types of vessels / columns such as stirred tank [13], slurry bioreactor [16], airlift [22,23,26], bubble columns [14,15,17,18,20,21,27,28] have

been used in mass transfer studies. In literature there have not been many reports on studies relating to the volumetric mass transfer coefficient for oxygen in biodegradation of phenol in bubble column bioreactors. In this study the effect of feed concentration, air flow rate and feed flow rate on the oxygen mass transfer coefficient has been studied.

2 Materials & methods

Experimental set-up

The experimental set-up is shown in the Fig. 1. The bubble column bioreactor is made of glass. A sparger made of glass has been provided at the bottom of the reactor through which air can be sparged into the reactor. The active volume of the reactor is about 3.54 liters. The top of the glass reactor is closed with a plate through which all the probes and sensors are inserted into the reactor. An overflow line has been provided at the top so that, the reaction medium flows out of the reactor in continuous operation. The reactor is provided with a glass jacket to control the temperature of the reactor using a cooling / heating medium. To maintain the pH of the system a pH meter and a controller have been provided. Oxygen will be consumed in the degradation of phenol by microorganism. Oxygen required for the process was supplied in the form of air from a compressor. The flow rate of air was measured using rotameter, with a range of 1–10 lpm (liter/min).

2.1 Biodegradation Studies

Culture Preparation

Pseudomonas putida (NCIM-2650) reported to be capable of using phenol as carbon source, has been collected from National Collection of Industrial Microorganisms (NCIM) of National Chemical Laboratory (NCL), Pune, India. The culture was maintained by periodic subculture on nutrient agar and stored in a refrigerator. The reaction medium was prepared from this strain by growing the bacteria on 3.54 liters of 50 ppm of phenol solution containing growth medium. The composition of the growth medium is KH_2PO_4 420 mg/l, K_2HPO_4 375 mg/l, $(NH_4)_2SO_4$ 240 mg/l, NaCl 15 mg/l, $CaCl_2$ 15 mg/l, $MgSO_4.H_2O$ 30 mg/l. Sterilization of the phenol solution was done before inoculation of the organism. This has been done to selectively grow the microorganism. After the inoculation, the bacteria was allowed to grow in incubator at 30°C for

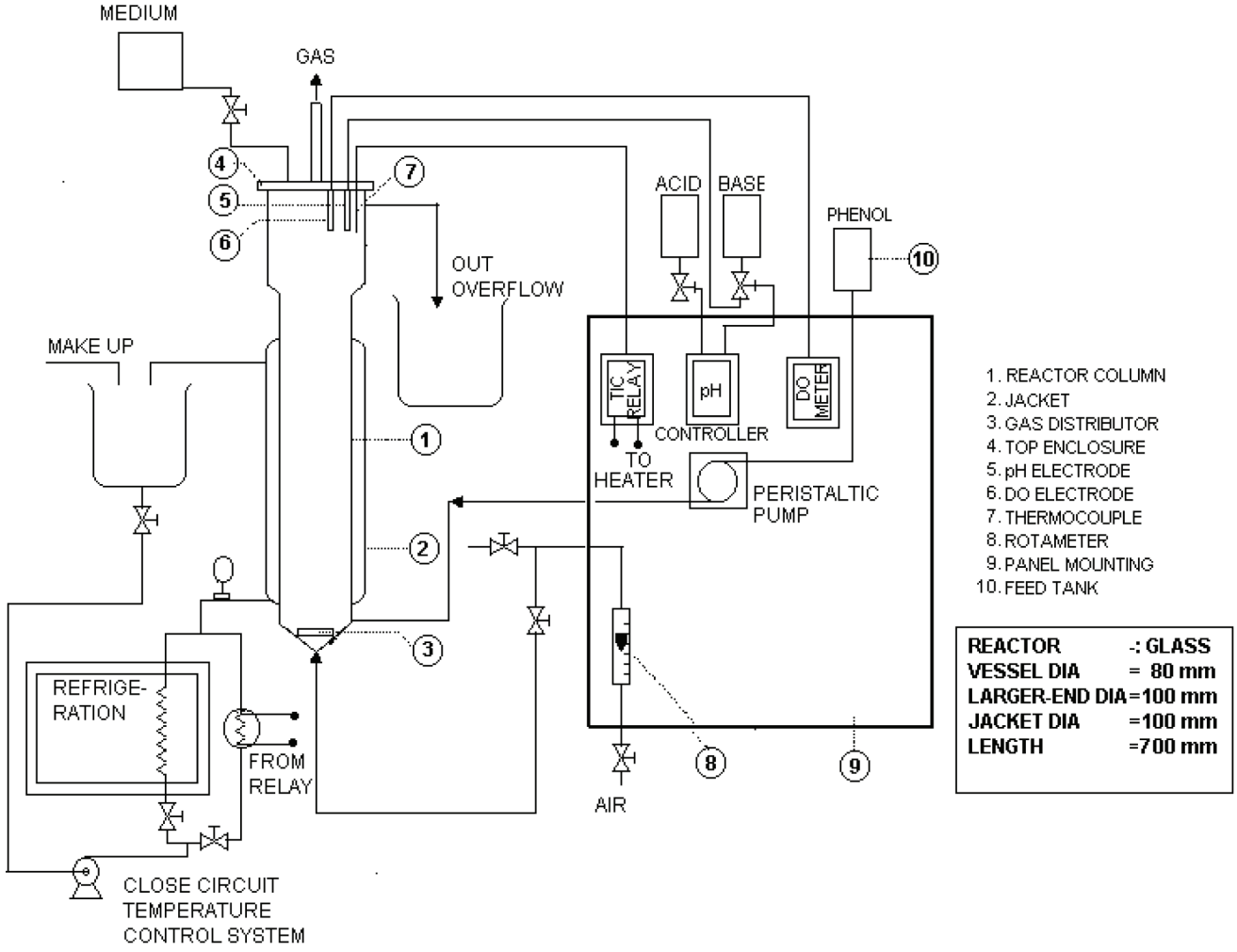

Fig. 1. Schematic of the experimental set-up

Tab. 1. Mass transfer correlations

Correlation	Column type	Reference
$\frac{k_L a T^2}{D_L} = 1.41 \cdot 10^{-3} \cdot \left(\frac{\mu_g}{\rho D_L}\right)^{0.5} \left(\frac{T^2 N \rho}{\mu_g}\right)^{0.67} \left(\frac{\rho N^2 T^2}{\sigma}\right)^{1.29}$	Stirred Tank	Albal et al [13].
$k_L a = 0.452 \left(\frac{D}{D_c^2}\right) Sc^{1/2}\, Re^{3/4}\, Fr^{7/60} Bo^{3/5}$	Bubble column	Kawase et al [14].
$Sh = 0.62 Sc^{0.5} Bo^{0.33} Fr^{0.68} \left(\frac{\rho_g}{\rho_l}\right)^{0.04}$	Bubble Column	Ozturk et al [15].
$k_L a = 7CF\sqrt{D_A}\,\frac{u_g \rho_L^{17/20}\sqrt{g}}{\mu_B^{1/4}\sigma^{0.6} d_c^{1/6}}$	Slurry Bioreactor	Kawase and Moo-Young [16].
$k_L a_L = 2.39 \times 10^{-4} \left(\frac{P_G}{V_L}\right)^{0.86}$	Bubble column bioreactor	Rubio et al [17].
$k_L a = K \times 10^{-3.08} \left(\frac{D_T V_g \rho_g}{\mu_l}\right)^{0.254}$	Bubble Column	Kang et al [18].
$Sh = 1.76 \left(Sc\right)^{0.5} \left(Re_j\right)^{0.99} \left(Fr\right)^{0.53} \left(Mo\right)^{-0.06} \times \left(\frac{0.15 + 1.05 L_d}{H_n + L_d}\right)^{0.71}$	Jet-loop bioreactor	Jamshidi et al [19].
$k_L a = \left(2 + f \cdot \frac{2}{\sqrt{\pi}} \cdot \sqrt{\frac{w_b \cdot d_b}{D_{o2}}}\right) \cdot \frac{D_{o2}}{d_b} \cdot \frac{6 \cdot \varepsilon}{d_b}$	Bubble Column Bioreactor	Weuster-Botz et al [20].
$k_L = 0.448 \left(\frac{P/V}{\rho}\right)^{1/4} \left(\frac{D_L}{r}\right)^{1/2}$	Bubble column	Linek et al [21].
$k_L a = 0.531 \mathrm{J}_{GR}^{0.762}$ for packed bed At only low superficial gas velocities JGR (<0.006 m/s), $K_L a$=2.530 J_GR-0.003 with packing $K_L a$=0.7369 J_GR-0.00005 without packing	external loop airlift bioreactor	Nikakhtari and Hill [22].
$k_L a = 1.552 \left[u_{gr}\right]^{0.935} \left[v\right]^{-0.683}$	draft tube airlift bioreactor	Shariati et al [23].
$k_L a = 0.014 u_G^{0.66} \mu^{-0.5}$	Bubble column	Dhaouadi et al [24].
$k_L a = 1.45 \left\{1 - \left(\frac{X}{1.07 \times 10^4}\right)^{2.55}\right\} U_g^{0.75}$	Slurry bubble column	Mineta et al [25].
$Sh = 4.6 \times 10^{-5} Fr^{0.642} Sc^{0.779} Ga^{0.673} Bo^{0.245} \varepsilon^{0.200}$	Airlift bioreactor	Cerri and Badino [26].
$k_L a = a_1 u_G^{a_2} \left(1 + d_p\right)^{a_3} \left(1 - e_S\right)^{a_4}$	Bubble Column	Mena et al [27].
$k_L a = 3.372 \times 10^3 \sqrt{\frac{D_L}{\pi}\sqrt{\frac{u_G g}{\mu_l}} u_G^{0.87} \mu^{*-0.24}}$	Bubble Column	Ferreira et al [28].

24 hours. The bacteria was subcultured once in a month by preparing slants using nutrient agar of composition (for 100 ml of nutrient broth/agar): Beef Extract 1.0 g, NaCl 0.5 g, Peptone 1.0 g, agar-agar 2.0 g. Sterile conditions were not maintained during the continuous operation of the reactor. Studies have been carried out at feed concentrations of 50, 100, 150, 200 and 250 mg/l, feed flow rates of 390, 450, 510, 570, 630 ml/h and air flow rates of 1, 2, 3 and 4 lpm. Temperature and pH were maintained at 30°C and 7 respectively. Phenol concentration has been determined using iodometric method [29].

2.2 Dynamic method for measuring oxygen mass transfer coefficient [30]

The dynamic method for k_La determination is based on the response of the dissolved oxygen concentration to changes in the inlet gas phase oxygen concentration. This technique employs the liquid phase oxygen balance equation. The dynamic method used has the advantage compared to the other method available in literature viz., static method, that prior knowledge of the flow behavior of the gas is not required. Oxygen balance gives

$$\frac{d\left(C_{O_2}\right)}{dt} = \frac{F\left(C_{O_2,in} - C_{O_2}\right)}{V_L} + k_La\left(C^*_{O_2} - C_{O_2}\right) - r_{O_2} \quad (1)$$

From literature [5,7,8] it may be noted that the values of k_La are in the range of 0.002 to 0.016 s^{-1}. The term F/V_L in the present work, is very small in comparison to k_La. Therefore the above equation may be simplified to

$$\frac{d\left(C_{O_2}\right)}{dt} = k_La\left(C^*_{O_2} - C_{O_2}\right) - r_{O_2} = k_La\left(C^*_{O_2} - C_{O_2}\right) - q_{O_2}X \quad (2)$$

The equation can be used to determine k_La by first halting aeration to fluidized bed bioreactor (Fig. 2). Concentration of dissolved oxygen is measured using a DO meter. If the gas phase disengages quickly from the liquid and there is no surface aeration, then transport term disappears from the above equation and it reduces to

$$\frac{d\left(C_{O_2}\right)}{dt} = -q_{O_2}X \quad (3)$$

where $q_{o_2}X$ is the microbial volumetric rate of oxygen consumption. If non-gassing period is short, the microbial suspension will continue to respire at the same rate and dissolved oxygen will decrease linearly with time. q_{o_2} is assumed to be independent of C_{o_2}. $q_{o_2}X$ can be obtained from the non-gassing period. Rearrangement of equation (2) gives the following equation:

$$\begin{aligned}\frac{d\left(C_{O_2}\right)}{dt} &= k_La\left(C^*_{O_2} - C_{O_2}\right) - q_{O_2}X \\ &= k_La\left\{\left[C^*_{O_2} - \frac{q_{O_2}X}{k_La}\right] - C_{O_2}\right\}\end{aligned} \quad (4)$$

The above equation upon integration gives

$$ln\frac{\left[C^*_{O_2} - \frac{q_{O_2}X}{k_La}\right] - C^{t=0}_{O_2}}{\left[C^*_{O_2} - \frac{q_{O_2}X}{k_La}\right] - C_{O_2}} = k_Lat \quad (5)$$

After reestablishment of steady state dissolved oxygen concentration is given by

$$C^*_{O_2} - \frac{q_{O_2}X}{k_La} = C^{t=\infty}_{O_2} \quad (6)$$

Using equation (6), equation (5) can be modified to

$$ln\frac{C^{t=\infty}_{O_2} - C^{t=0}_{O_2}}{C^{t=\infty}_{O_2} - C^{t}_{O_2}} = k_Lat \quad (7)$$

In the plot of log term against time, slope of the straight line gives mass transfer coefficient k_La.

3 Results and discussion

Volumetric mass transfer coefficient (k_La) for oxygen was determined at feed concentrations of 50, 100, 150, 200 and 250 mg/l, feed flow rates of 390, 450, 510, 570 and 630 ml/h and air flow rates of 1, 2, 3 and 4 lpm. Since oxygen is sparingly soluble in liquid phase (wastewater) compared to gas phase, the resistance of the gas phase was neglected. The results of the study are shown in the Figs. 3 – 12. The effect of feed flow rate at various air flow rates is shown in Figs. 3 – 7. From the figures it can be observed that, for a given feed flow rate, as the air flow rate is increased the oxygen mass transfer coefficient increases. Feed flow rate has relatively insignificant effect. Increasing the feed flow rate, for a given air flow rate, resulted in only marginal increase in the mass transfer coefficient. The values of the k_La have been found to be in the range of 0.00513 – 0.01793 s^{-1}. These values give an idea of resistance to oxygen mass transfer from gas phase to liquid phase in the biodegradation of phenol. There have been reports in literature relating to oxygen mass transfer coefficient on phenol biodegradation. Worden and Donaldson [31] in their study on dynamics of a fluidized bed bioreactor (FBR) treating phenol, obtained K_La (overall coefficient) in the absence of reaction in the range 0.005 – 0.01 s^{-1}. They used deoxygenated water in the experiments to transfer oxygen from gas phase to liquid (water) phase. In another dynamic study on phenol degradation in FBR, Tang et al. [32] have used a value of 0.0139 s^{-1}. Venu Vinod and Reddy [33] have reported oxygen mass transfer coefficient values in the range of 0.0039 – 0.0139 s^{-1}. It can be seen that the mass transfer coefficient values obtained in FBR are smaller than those in bubble columns. In dynamic method it is assumed that F/V_L is negligible compared to k_La. In the present work that value of F/V_L is of the order of 10^{-5} s^{-1}, which is very small compared to the values of the mass transfer coefficient (k_La). Therefore the assumption is justified.

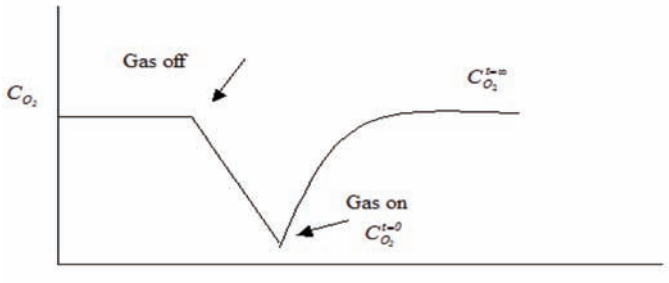

Fig. 2. Typical DO concentration profile using dynamic method for the determination of $k_L a$

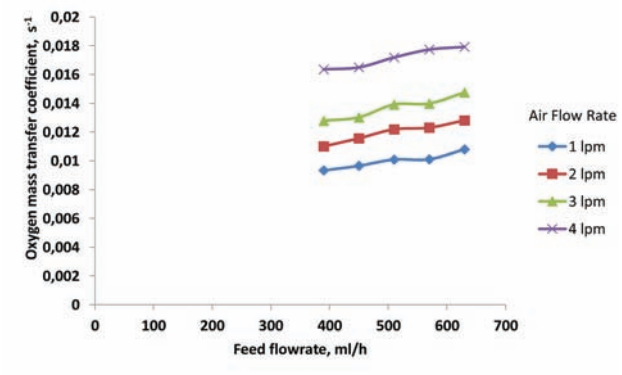

Fig. 3. Variation of $k_L a$ with feed flow rate and air flow rate (feed concentration 50 ppm)

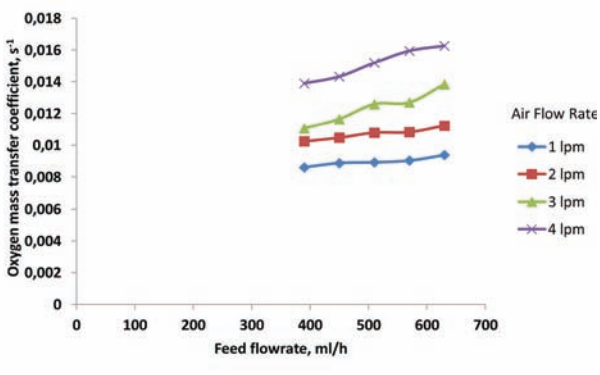

Fig. 4. Variation of $k_L a$ with feed flow rate and air flow rate (feed concentration 100 ppm)

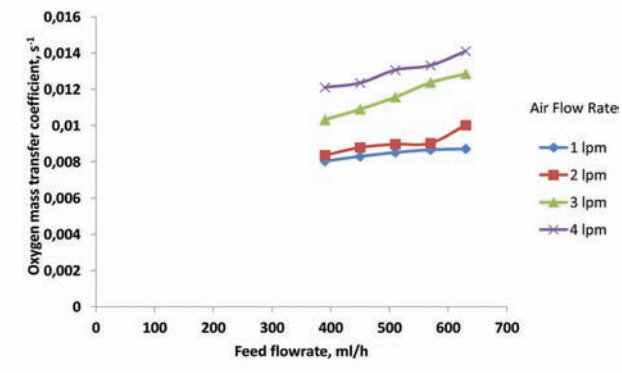

Fig. 5. Variation of $k_L a$ with feed flow rate and air flow rate (feed concentration 150 ppm)

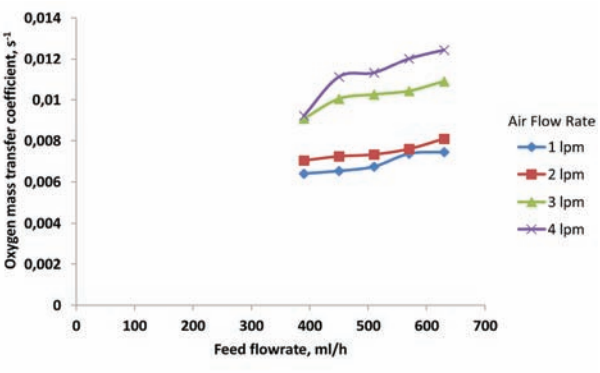

Fig. 6. Variation of $k_L a$ with feed flow rate and air flow rate (feed concentration 200 ppm)

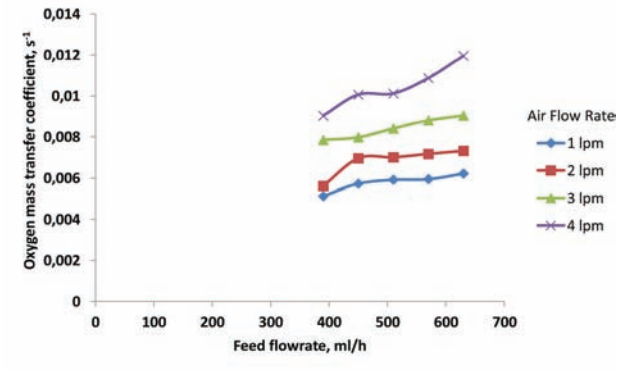

Fig. 7. Variation of $k_L a$ with feed flow rate and air flow rate (feed concentration 250 ppm)

The effect of feed concentration on the mass transfer coefficient at various air flow rates has been shown in Figs. 8 – 12. As the feed concentration increases the oxygen mass transfer has been found to decrease. This is due to the fact that at higher feed concentrations, the reaction medium was more viscous due to the presence of higher concentration of biomass in the bioreactor thereby decreasing the solubility of the oxygen. This results in lower mass transfer coefficients.

Dimensionless correlation for mass transfer coefficient:

There are no correlations in literature for k_La for biodegradation of phenol in a bubble column bioreactor. In this work a correlation has been developed in terms of dimensionless numbers Bond number (*Bo*), Galileo number (*Ga*), Froude number (*Fr*) and Schmidt number (*Sc*). One correlation is based on the bubble column diameter and the other is based on the diameter of the bubble at the orifice. The volumetric mass transfer coefficient is dependent on the gas velocity (U_g), the diameter of the bubble at the orifice (D_b), properties of the liquid medium in the bioreactor such as density (ρ_l), viscosity (μ_l) and surface tension (σ_l), and diffusivity of oxygen in water (D_{ow}). Bubble diameter was calculated using Moo-Young and Blanch equation [34]

$$D_b = 0.19 D_o^{0.48}\, Re_o^{0.32}$$

where D_o is the diameter of the orifice (on sparger through which air is sparged into the bioreactor) and Re_o is the Reynolds number for air flow at the orifice. Viscosity (μ_l) and surface tension (σ_l) were determined using Canon-Fenske viscometer and stalagmometer respectively. Diffusivity was calculated using the Wilke-Chang equation [35].

$$k_La = f\left(U_g, \rho_l, \mu_l, \sigma_l, D_b, g, D_{ow}\right)$$

The variables can be combined in the form of dimensionless groups as

$$Sh = f\left(Bo_l, Ga_l, Fr_l, Sc_l\right)$$

Sherwood number, $Sh = \dfrac{k_L a D_b^2}{D_{ow}}$

Bond number, $Bo_l = \left(\dfrac{g\rho_l D_b^2}{\sigma_l}\right)$

Galileo number, $Ga_l = \left(\dfrac{g D_b^3 \rho_l^2}{\mu_l^2}\right)$

Froude number, $Fr = \left(\dfrac{U_g^2}{D_b g}\right)$

Schmidt number, $Sc_l = \left(\dfrac{\mu_l}{\rho_l D_{ow}}\right)$

The values of Bond number, Galileo number, Froude number, Schmidt number and Sherwood number are in the range of 188 – 211, 3.37×10^8 - 1.73×10^9, 3.06×10^{-5} - 3.17×10^{-4}, 402 -831 and 0.39×10^4 – 2.8×10^4 respectively. The developed correlation is

$$Sh = 0.455\left(Bo\right)^{-0.547}\left(Ga\right)^{0.82}\left(Fr\right)^{0.489}\left(Sc\right)^{0.5}$$

The equation fits the experimental data well as indicated by the high value of correlation coefficient (R^2=0.964). A comparison between experimental data and predicted values of k_La is shown in Fig. 13. It can be seen that the correlation predicts almost all the experimental data within 17.5% error.

There are a large number of correlations published for k_La for various systems. A comparison of the values of k_La predicted by the correlation developed in the present work and correlations reported in literature is shown in the Fig. 14. Correlations have been developed for air – water systems [22,24,36,37]. Different liquids have been used by researchers for evaluating the volumetric oxygen mass transfer coefficient, such as aqueous solutions of glycerol, methanol and sodium sulphite [36] non-Newtonian fluids such as solutions of carboxymethyl cellulose (CMC), poly acrylamide (PAA) and xanthan gum [38] aqueous solutions of carboxypoly methylene and CMC [14], aqueous sucrose solution [39] and aqueous NaOH solution [40]. Unlike the correlations for gas holdup where many of the correlations involve only gas velocity, dimensionless correlations for k_La (Sherwood number) have been reported in terms of dimensionless numbers such as Schmidt number, Bond number, Galileo number and Froude number. Few correlations have been published considering only gas velocity [22,40] and gas velocity and liquid viscosity [24]. Fig. 14 shows only the values which are close to the values in the present study. Other correlations [15,41-43] from literature have been used to predict the values of k_La, but the predictions are not close to the values of the present study, and therefore not shown in the figure. There are hardly any studies reported on mass transfer in bubble column bioreactors using actual biological systems. Mineta et al. [25] have studied mass transfer in a dense activated sludge slurry bubble column degrading *p*-nitrophenol and developed a correlation for k_La in terms of gas velocity (U_g) and waste activated sludge concentration (X), which is given below:

$$k_La = 1.45\left\{1-\left(\frac{X}{1.07\times10^4}\right)^{2.55}\right\}\mathrm{U}_{\mathrm{g}}^{0.75}$$

This equation is not suitable to predict the value of k_La in the present study, as the X (biomass concentration) values in the present study are very small compared to those of Mineta et al. [25]. As the correlations for biological systems are scarce, the correlation developed in the present work addresses this gap, as it is developed using a biological system and considers all the variables affecting the mass transfer coefficient.

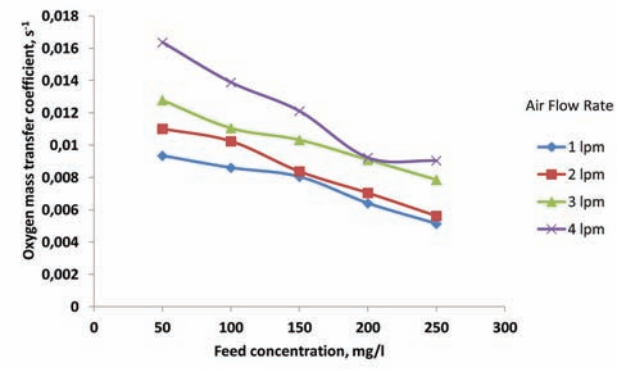

Fig. 8. Variation of k_La with feed concentration and air flow rate (feed flow rate 390 ml/h)

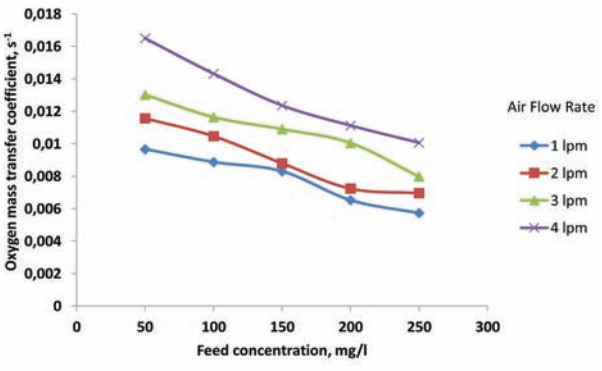

Fig. 9. Variation of k_La with feed concentration and air flow rate (feed flow rate 450 ml/h).

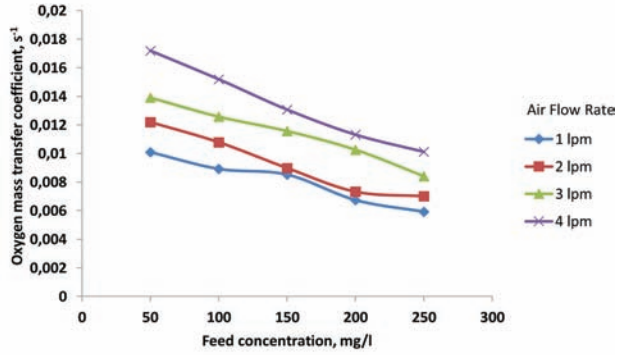

Fig. 10. Variation of k_La with feed concentration and air flow rate (feed flow rate 510 ml/h)

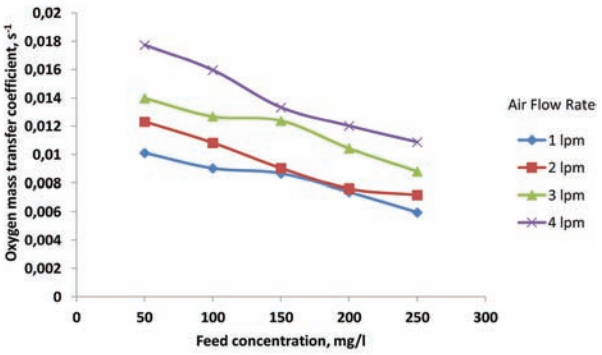

Fig. 11. Variation of k_La with feed concentration and air flow rate (feed flow rate 570 ml/h)

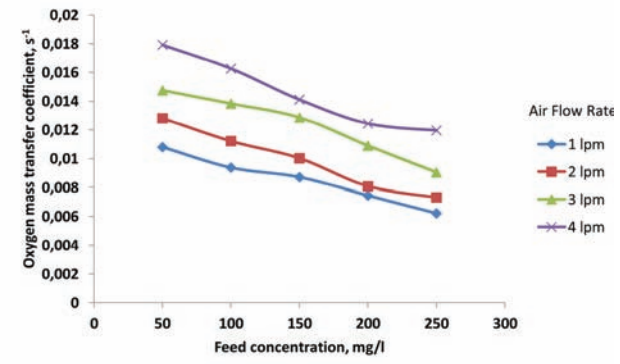

Fig. 12. Variation of k_La with feed concentration and air flow rate (feed flow rate 630 ml/h)

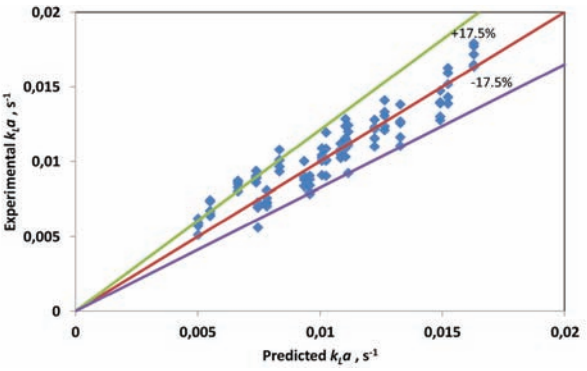

Fig. 13. Comparison of experimental and predicted k_La.

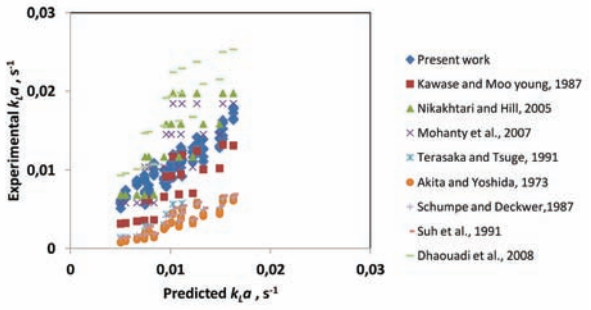

Fig. 14. Comparison between the experimental k_La and predicted k_La from correlations in literature.

4 Conclusions

Volumetric mass transfer coefficient has been determined for biodegradation of phenol in bubble column bioreactor using the dynamic technique. Volumetric oxygen mass transfer coefficient is dependent on superficial gas velocity, liquid properties and feed concentration. Feed flow rate did not have much effect on k_La. A mathematical correlation is derived and presented for k_La in terms of dimensionless numbers. Bubble columns give greater values of oxygen mass transfer coefficient than fluidized bed bioreactors.

Nomenclature

a :Interfacial area per volume (m^2/m^3).

Bo_l :Bond number = $\left(\frac{g\rho_l D_b^2}{\sigma_l}\right)$.

C_{o_2} :Dissolved oxygen concentration in the reactor (kg/m^3).

$C^*_{o_2}$:Dissolved oxygen concentration under equilibrium conditions with air (kg/m^3).

$C_{o_2,in}$:Dissolved oxygen concentration in the feed to the reactor (kg/m^3).

D_0 :Orifice diameter (m).

D_b :Bubble diameter (m).

D_{ow} :Diffusion coefficient of oxygen in water (m^2/s).

F :Volumetric flow rate of feed (ml/h).

Fr_g :Froude number = $\left(\frac{U_g^2}{D_b g}\right)$.

G :Acceleration due to gravity (m/s^2).

Ga_l :Galileo number = $\left(\frac{gD_b^3\rho_l^2}{\mu_l^2}\right)$.

k_La :Volumetric oxygen transfer coefficient (s^{-1}).

lpm :liter/min.

q_{o_2} :Specific rate of microbial oxygen consumption (1/h).

r_{o_2} :Volumetric rate of oxygen consumption (kg/m^3-h).

Re_0 :Orifice Reynolds number.

Sc_l :Schmidt number = $\left(\frac{\mu_l}{\rho_l D_{ow}}\right)$.

Sh :Sherwood number = $\left(\frac{k_l a D_b^2}{D_{ow}}\right)$.

V_L :Liquid volume in reactor (m^3).

U_g :Superficial gas velocity, Gas velocity (m/s).

X :Activated Sludge Concentration (mg/l, kg/m^3).

Greek letters

μ_l :Viscosity of effluent (kg/m.s).

ρ_l :Density of effluent from bioreactor (kg/m^3).

σ_l :Surface tension (N/m).

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