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Mass Transfer Coefficient ($k_L.a$) Determination with Microelectrodes in Biofilms From an RBC at Different Operation Conditions

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Abstract

The mass transfer coefficient of oxygen ($k_L.a$) was determined for biofilms produced in a rotating biological contactor (RBC) by means of a microelectrode built at the laboratory. Three different rotational velocities (1.25, 2.75 and 5.25 rpm) were used for the determination. The microelectrode tip was made of platinum (99% purity); which was mechanically filed down from its initial diameter of 0.5 mm to 67 μm. The microelectrode was then mounted on a micromanipulator with a precision of 100 μm. The RBC was fed with municipal wastewater, reaching a removal efficiency of 65% as soluble COD and 96% removal of nitrogen as ammonia nitrogen ($N\text{-}NH_4$). The hydraulic retention time (HRT) was 8 h. The average room temperature was 26.7 °C and the water temperature in the RBC was constant (22 °C) during the measurement. The $k_L.a$ determined at 1.25 rpm was 1.953 $(day)^{-1}$, while that determined at 2.75 rpm was 3.117 $(day)^{-1}$ and finally at 5.25 rpm was 9.799 $(day)^{-1}$. The results confirmed the irregularity of the biofilm's structure.

Keywords: *Mass transfer, coefficient, microelectrode, biodisk, biofilms.*

1. Introduction

Rotating biological contactors (RBC) have been used to eliminate pollutants present in municipal wastewaters, as well as other specific organic compounds like azo dye mixtures [1,2] and 2,4-diclorophenol, among others. Their use is justified since they are economical and almost all liquid effluents contain a large amount of biodegradable material. An RBC has a very high interfacial area, being virtually independent of rotational velocity, which is an advantage over other biological treatments of wastewaters. Additionally, they can be easily adapted to small and medium wastewater treatment plants, with easy operation and maintenance [3].

However, the aerobic biodegradation is frequently limited by oxygen diffusion in the biofilm [4,5]. The depth to which oxygen penetrates into the biofilms depends on different factors such as density, composition of the biofilm and angular velocity [1,2]

Different studies on mass transport in biofilms have been conducted. It has been shown that biofilms form cellular clusters separated by interstitial voids filled with water and biopolymers[8,9]. Moreover, due to the irregular distribution of biomass in the biofilms, the mass transfer is affected and the effective diffusivity varies across the biofilm[10]. On the other hand, it has been demonstrated that the density of the biofilms depends on its depth, being higher at the bottom of the biofilms[11]. Several researchers have directly applied microelectrodes to measure the oxygen concentration and to obtain the profiles of oxygen in the biofilms. In another research[12,13], a platinum microelectrode was used to determine the oxygen concentration in the biofilm; it was concluded that the liquid flow in the biofilm is affected by irregularities in its surface. Fu et. als [14] used a method of finite differences to determine the effective diffusivity in a completely mixed bioreactor; the determination was carried out in stable state, using a microelectrode at intervals of 100 μm. It was concluded that the effective diffusivity of the oxygen is highly related to the density of the biofilm. Furthermore, different microelectrodes have been used to measure the oxygen concentration of ammonia nitrogen, nitrates and pH[15,16]. In another study, a platinum-tipped microelectrode was used to determine oxygen flows within the biofilms of an RBC [17].

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Yang and Lewandowski[18] calculated the local mass transfer coefficient in the biofilm formed in an open channel biofilm reactor using synthetic water. In this study, a modified limiting current technique was applied. It was concluded that the mass transfer coefficient is not constant, but it varies both horizontally and vertically.

Another study [19,20] determined that the oxygen transfer coefficient ($k_L.a$) varied according to different percentages of immersion of the disks and different rotational velocities, thus theoretically determining the influence of the body of water upon the $k_L.a$. In this study, the $k_L.a$ is 50 % less than the values theoretically calculated. It is also proposed an empirical correlation to calculate the $k_L.a$ as a function of the velocity of rotation, as well as the percentage of immersion and the geometrical dimensions.

Kubsad et al. [4] proposed a model (modified Kim and Molof) to determine $k_L.a$ using an RCB without biomass growth. This model showed a good correlation with the significant physical parameters.

The objective of this paper is to determine the local $k_L.a$ of the oxygen profiles obtained from the biofilm of a laboratory scale RBC at different rotational velocities (1.25, 2.75 and 5.25 rpm). The determination was carried out by means of a microelectrode at different biofilm depths. It was indirectly determined the necessary oxygen for the oxidation of nitrogen ammonia present in a municipal wastewater influent.

2. Material and methods

The rotating biological contactor (RBC) used in this study was made of 20 acrylic disks. The tub and the primary settler of the RBC were built of fiberglass; the form of the tub was a hollow semicylinder placed in a metallic structure. The reactor tub was divided into five equal-sized transverse compartments using fiberglass screens. The reactor's chambers were interconnected through a lateral slot located on the upper part of each screen. The slots were arrayed in zigzags, allowing the pass of both liquid and microbial flocculate toward the next chamber when the maximum capacity level had been reached (Fig. 1).

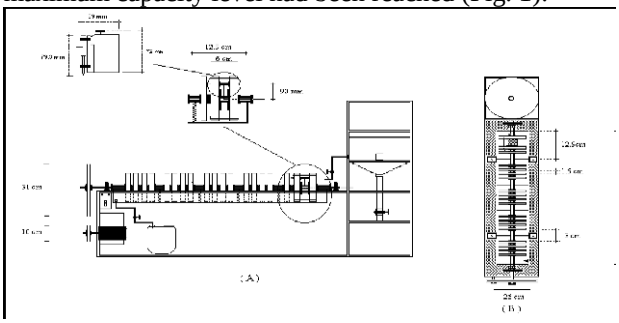

Figure 1. Configuration scheme rotating biological reactor (RBR).

Thus, the biological degradation process is carried out sequentially in this system. Table 1 lists the characteristics of the RBC system.

Table 1: Characteristics of RBC used

Parameter	***Value***
Number of stages	5
Number of disks by stage	4
Diameter disks	0,241
Thick the disks	0.035
Surface area by stage	0..3804 m^2
The volume of water by stage	0.0022 m^3 (stage 1-4), 0.002(stage 5)
Area /volume ratio	1.9097 m^2/0.0108m^3 (176.122)
Percentage of immersed the disks	45 % (stage 1-4) and 40 % (stage 5)

Municipal wastewater was fed to the reactor with a variable-speed pump. It is to be noted that the chemical oxygen demand (COD) of the wastewater was not constant, but rather changed during the day.

It was followed the same procedure described in other studies [17,21,22] to build the microelectrode, with only the following modification [23,24]: the diameter of the tip was obtained by means of a Struers file model Dap-7, using Buehler silicon carbide sandpaper (5000 and 1200 mesh). The tip of the microelectrode was reduced until reaching a diameter of 67 μm.

The microelectrode was installed in the last stage. To keep a fixed distance between each disk, support axes were used to avoid perturbations during measurements. The microelectrode was attached to the micromanipulator, which was in turn attached to the disk. The micromanipulator had a displacement precision of 100 μm. A copper wire with a special covering to avoid any induced voltage was attached to the microelectrode, serving as wire conductor. This passed through the center of the disk and the RBC axis.

The determination of dissolved oxygen (DO) was carried out after four months of operation of the RBC. The DO measurements were carried out moving the microelectrode from the liquid into the biofilm in 100-μm increments for each rotational velocity (1.25, 2.75 and 5.25 rpm). Additionally, measurements were made in five points along the diameter of the disks as shown in the figure 2. After the DO measurements were taken at one point, the biodisks were allowed to rotate once again at the fixed rpm for 20 minutes, then the biodisk was stopped and the microelectrode was placed in another point to perform the DO measurements.

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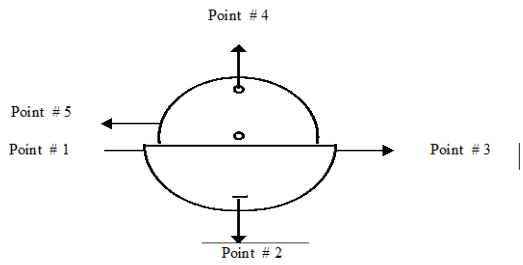

Figure 2. Points of disk where dissolved oxygen was determined

For each point and penetration distance, 20 data were taken for the determination of the relative standard deviation and mean. For relative standard deviation inferior to 0.5 the data would be accepted; the mean was taken as a representative for this point.

An Ag/AgCl commercial reference electrode was used. A digital multimeter was used to measure the current on the microampere (μA) scale. To carry out the microelectrode calibration curve, the oxygen concentration was plotted against current with a correlation of R^2 = 0.975. The oxygen concentration was determined from equation 1 with the electric current measured by means of the microelectrode [23].

$$C_{o_2} = -0.00291812 + 0.00001429 * \mu \quad (1)$$

Where:
CO_2 = concentration of oxygen (mg.cm^{-3})
μ = measured current (μA)

The oxygen concentration at top of the biofilm layer (biofilm depth = 0.0 cm) and into the biofilm at different depths was measured with the microelectrode for each test carried out at different rpm.

3. Results

The oxygen concentration profiles obtained at 1.25 rpm are shown in figure 3. The DO concentrations can be observed according to figure 2. The measurements made in the water in point 5 run from concentrations of 0.45 to 0.05 mg/L DO inside the biofilm (1500 μm).

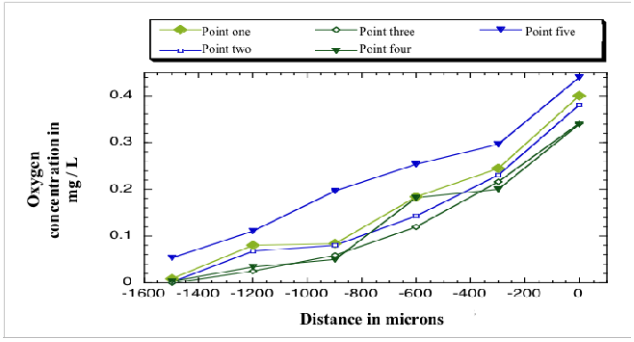

Figure 3. Oxygen concentration profiles in the RBR biofilms at 1.25 r.p.m.

The DO profiles at 2.75 rpm are shown in figure 4. At this rotation speed the DO concentration in the biofilm behaves similarly to that of 1.25 rpm; nevertheless, DO concentrations in the surface and inside the biofilm at different sampling points run from 0.8 to 1.6 mg L^{-1}, although the thickness of the biofilm is 60 % smaller.

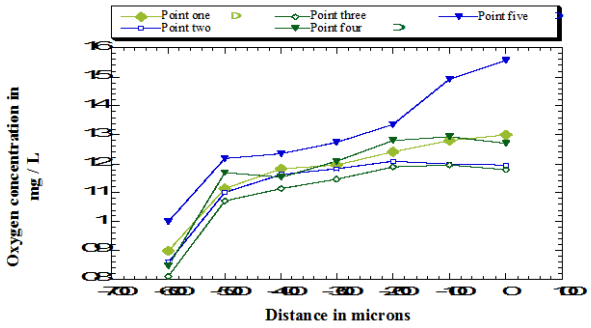

Fig. 4. Oxygen concentration profiles in the RBR biofilms at 2.75 r.p.m.

The DO profiles at 5.25 rpm are shown in figure 5. In this case, the profiles run from 0.05 to 2 mg/L. It was found that the biofilm's thickness was 1600 μm regarding the thickness measured at the 2.75 rpm test.

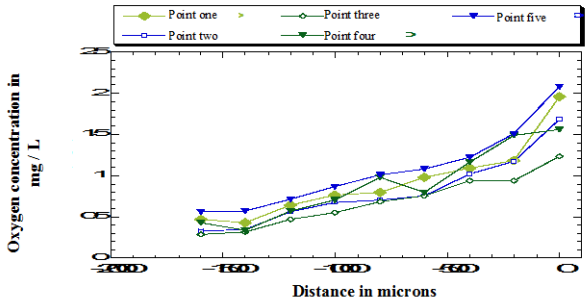

Fig. 5. Oxygen concentration profiles in the RBR biofilms at 5.25 r.p.m.

Comparing figures 3 to 5, they all exhibit a similar behavior. The oxygen diffusivity decreases as the biofilm

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depth increases. There is a maximum value in the biofilm's surface, but there is a drastic decrease in the DO concentration at the next point.

Oxygen flows were determined from the oxygen profiles through equation (2) proposed by Nishidome et al. [17] .

$$F_o = \frac{(C_o - C_{oL}) D_f}{L} \qquad (2)$$

Where:

F_O = Oxygen flow (g of O_2/m^2.d)

C_O = Oxygen concentration on the surface the biofilm (g/m^3)

C_{OL}= Oxygen concentration to a specific thickness (L) from the surface of the biofilm (g/m^3)

D_f = Oxygen molecular diffusion coefficient in the biofilm (m^2. day^{-1}) = $2.1x10^{-4}$ $m^2.day^{-1}$

L = Biofilm thickness from its surface (m)

In figures 6 can be observed that the soluble COD removal efficiency is much less than the $N\text{-}NH_4^+$ removal efficiency; therefore it can be said that the RBC's biofilm is delimited by nitrifying bacteria, upon not observing a significant oxidation of organic carbon. This shows that the oxygen consumption is mainly attributed to nitrification. Thus the oxygen flow can be determined by the multiplication of the ammonia oxidation —in this case at 5.25 rpm– by a stoichiometric reason of 4.569 g O_2 / g $N\text{-}NH_4$.

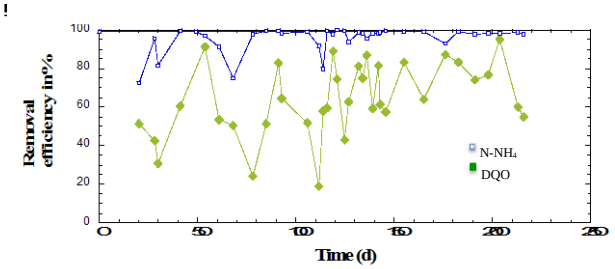

Fig. 6. Oxygen concentration profiles in the RBR biofilms at 5.25 r.p.m.

The oxygen flows were obtained at each penetration point of the biofilm and sampling point. The figure 7 shows the average flow values at three different rotational velocities.

This theoretical determination of oxygen flow was carried out only at 5.25 rpm as OD values in the first stage of the RBC tub at 1.25 and 2.75 rpm were below 1.0 mg/L. This brought adverse conditions for the biofilm's development.

The $k_L.a$ values were determined from the determined oxygen's flows through the equations proposed by Bitanja *et al.* (1975) (equation 3).

$$k_L.a = \left[F_o / (C_s - C_o) \right] * a \qquad \text{..... (3)}$$

C_s = oxygen saturation concentration in the residual water (mg of O_2/m^3)

C_O = concentration on the surface of the biofilm (mg of O_2/m^3)

F_O = average oxygen flow (mg of O_2/m^2.d)

$k_L.a$ = oxygen transfer coefficient (d^{-1})

a = area interphase by volume unit (m^{-1})

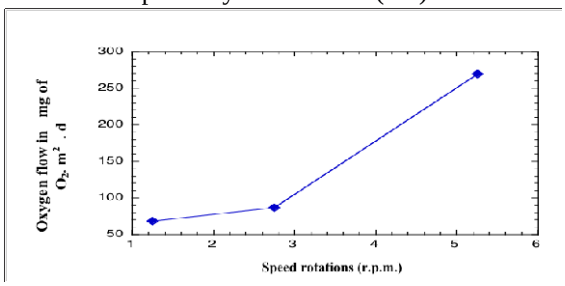

Fig. 7. Oxygen flow values at three different rotational velocities.

Figure 8 shows the $k_L.a$ behavior at the three rotational velocities used. The coefficient value at 5.75 is 8.121 d^{-1}, greater than at 1.25 (1.969 d^{-1}) and 2.75 (2.897 d^{-1}) rpm. This follows a similar behavior to that reported by Zeevalkink et al. [19], with a difference of 10 % less.

The last point of figure 9 is the value corresponding to 6.583 d^{-1} of $k_L.a$ at 5.25 rpm, but obtained in an indirect fashion. This value is 19 % less than the obtained experimentally through oxygen flows determined with microelectrodes.

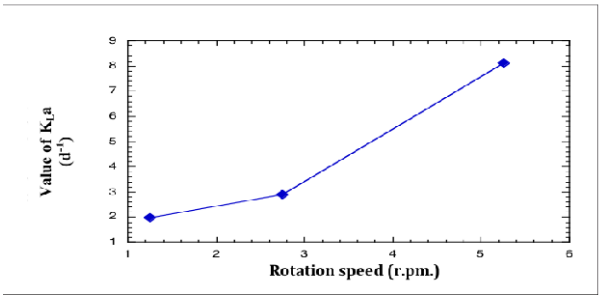

Fig. 8. K_La values at three different speeds of rotation

4. Conclusions

The oxygen flow data in the biofilms show significant irregularities in these structures, with cellular clusters directly affecting oxygen transport towards the inside of the biofilms.

Results show that the microelectrode technique is adequate for the $k_L.a$ determination in RBC biofilms due to all dissolved organic matter oxidation processes take place inside the biofilms. The $k_L.a$ is directly affected by the rotational velocities of the biodisks, which is confirmed

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when observing oxygen flow data as a function of the rotational velocities.

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