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# Oxygen Mass Transfer in Biological Treatment System in the Presence of Non-Aqueous Phase Liquid

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## Abstract

Oxygen mass transfer is an important phenomenon in any biological treatment systems, fermentation reactors and other biochemical reactors. In biological treatment processes, aeration and agitation is carried out to enhance the oxygen mass transfer in the system, which in turn enhances the fermentation or treatment efficiency. In case of wastewater treatment plant, this process takes around 45-50% of the total energy requirement of the treatment plant. This paper proposes a new method for enhancing an oxygen mass transfer phenomenon in biological treatment processes by introducing non-aqueous phase liquids in the treatment system.

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## 1. Introduction

Oxygen mass transfer is a critical and important phenomenon in any biological wastewater treatment systems, fermentation reactors and other biochemical reactors. In these aerobic processes, there is a thrust towards exploring new methods for supplying oxygen for attaining high cell concentration and reducing the energy requirement.

Mimura et al. (1969) first reported the effect of free phase hydrocarbon on oxygen transport in air-water hydrocarbon system [1]. Subsequently, various authors reported positive impact of different types of

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hydrocarbons in oxygen transfer phenomenon in fermentation processes, stirred tank bioreactors and rotating biological contactors [2-5]. In fermentation processes, it has been reported that oxygen mass transfer phenomenon can be enhanced by addition of non-aqueous phase liquids (NAPLs), like, hemoglobin, hydrocarbons and per-fluorocarbons [2]. These NAPLs are reported as oxygen vectors and offer advantages because of two reasons. Firstly, the solubility of oxygen in NAPLs is more than in water. It has been reported that the solubility of oxygen in C16 (hexadecane) was 8 times higher than in pure water and a mixture of hydrocarbons, from C12 (dodecane) to C16 was found to have oxygen solubility of 39.48 mg/L at 25°C [3, 6]. Secondly, these NAPLs have the capacity to carry oxygen from one phase to the other. It was found that oxygen mass transfer phenomenon in the presence of NAPL is a function of spreading coefficient ( $S_p$ ). When  $S_p < 0$ , the oil simply forms floating lens-like drops, which accumulates at the gas-liquid interface and oxygen transfer coefficient ( $K_{L,a}$ ) decreases linearly with increasing oil fraction. In contrast, when  $S_p > 0$ , oil spreads as a thin film on the gas-liquid interface and  $K_{L,a}$  increases with increasing oil fraction [7].

This paper proposes the utilization of these oxygen vectors for biological treatment processes to enhance the phenomenon of an oxygen mass transfer and reduce the energy requirement in conventional biological treatment processes. As per our knowledge, this is the first paper reporting the positive impact of NAPL (as oxygen vector) in biological wastewater treatment process.

## 2. Materials & Methods

Oxygen mass transfer experiments were carried out in Jar apparatus. Dissolved oxygen (DO) in the water was depleted with sodium sulfite. 40 mL of mixed liquor suspended solids (MLSS) was diluted to 800 mL by deoxidized water. Desired amount of oil was added to the solution. Paddle speed of the Jar apparatus was started at 75 rpm (kept constant) and simultaneously the stopwatch was started. DO was measured with DO probe (Eutech Instruments, Thermo Fischer Scientific Inc. USA) at regular intervals till saturation. Paddle speed as well as MLSS concentration was optimized before carrying out the experiments. Moreover, temperature and MLSS was measured. Linear regression was performed for determining  $K_{L,a}$  using SYSTAT 13.0. Moreover, three-parameter non-linear regression was performed to evaluate  $K_{L,a}$ ,  $C_s$  and  $C_o$  using SYSTAT 13.0. Three parameter non-linear regression analysis was utilized for determining these parameters based on  $C_t$  vs  $t$  profile using the equation:  $C_t = C_s - (C_s - C_o) \cdot \exp(-K_{L,a} \cdot t)$ , where,  $C_s$  = apparent saturation of gas in water, mg/L,  $C_o$  = initial concentration of oxygen in liquid, mg/L,  $C_t$  = concentration of oxygen in liquid at time  $t$ , mg/L.

In addition, oxygen uptake rate (OUR) was also calculated for two different types of sample. 300mL sample is taken from the experimental solution in a biochemical oxygen demand (BOD) bottle. It was stirred with the help of magnetic stirrer for one day to achieve DO saturation. DO meter was inserted in BOD bottle and the bottle was sealed with parafilm. Precautions were taken to ensure no bubble formation in the bottle. DO reading was noted down with respect to time. A graph of DO vs time was plotted which comes out to be a straight line with negative slope. From this slope, OUR was calculated.

## 3. Results & Discussion

Soybean oil was selected for carrying out the oxygen mass transfer experiments, as it was reported that soybean oil resulted in maximum oxygen transfer efficiency in fermentation process. It can be depicted from Fig. 1. Moreover, it was found to be more cost-efficient compared to other NAPLs. Initial experiments were performed to obtain optimum paddle speed and MLSS concentration in the absence of oil. At low paddle speed (<75 rpm), proper mixing was not observed and oxygen mass transfer rate was very slow and higher paddle speed (>75 rpm) resulted in in-consistent readings. Thus, 75 rpm was considered as optimum for

carrying out these experiments. Similarly, at higher MLSS concentration (2000-3000 mg/L), very slow oxygen mass transfer rate and in-consistent readings were observed. Lower MLSS concentration in the range of 150-200 mg/L was found to provide consistent readings. Thus, all subsequent experiments were carried out at paddle speed of 75 rpm and MLSS concentration of 150-200 mg/L.

Oxygen mass transfer experiments were carried out at different soybean concentrations: 0.01-0.75% (v/v). In clean water system, where  $C_s$  is known, a plot of  $\ln[(C_s - C_o)/(C_s - C_t)]$  vs  $t$  is typically used to determine  $K_L a$ . This approach could not be used in this three-phase system, since it is likely that  $C_s$  in the NAPL-water-biomass system may deviate from that in clean water system. This was confirmed by the poor fits obtained with one-parameter ( $K_L a$ ) non-linear regression of the re-aeration profiles ( $C_t$  vs  $t$ ). Thus, non-linear regression was performed with  $K_L a$ ,  $C_o$  and  $C_s$  as fitting parameters. Fig. 2 shows the fitting for linear and three parameter non-linear regression with 0.05% soybean oil concentration.

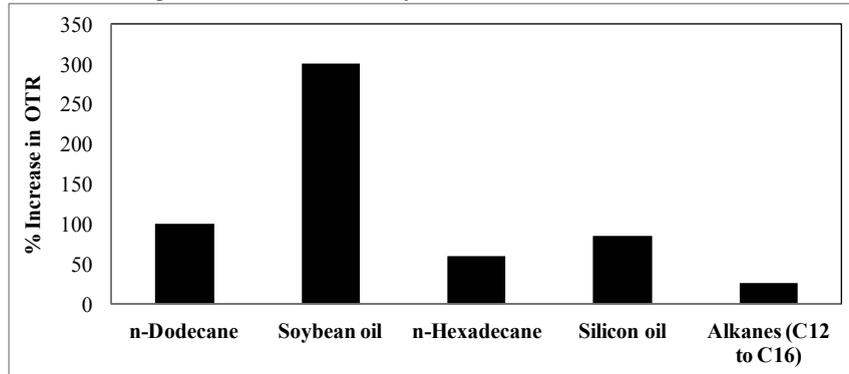


Fig. 1. Percent increase in oxygen transfer rate (OTR) with different types of NAPLs used by various researchers

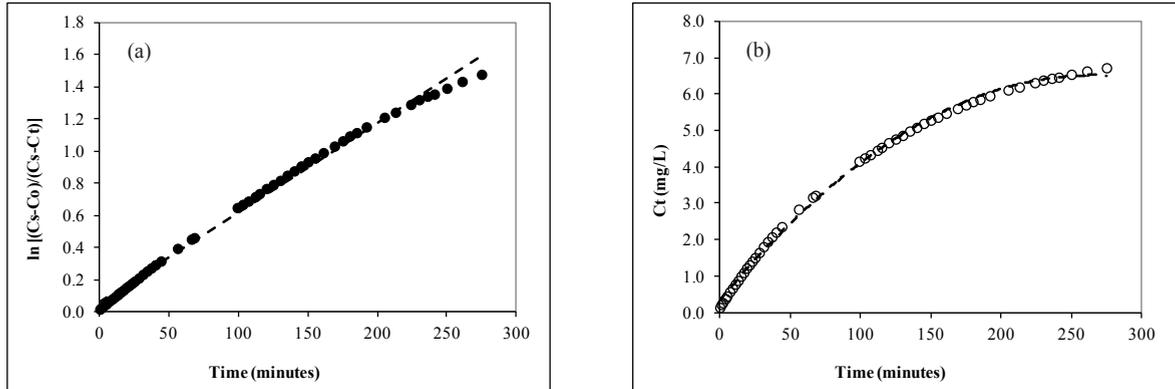


Fig. 2. Oxygen mass transfer in the presence of 0.05% soybean oil using: (a) linear regression and (b) three parameter non-linear regression

Considering three-parameter non-linear regression, the  $K_L a$  value increased from  $0.007$  to  $0.009 \text{ min}^{-1}$  with increased soybean oil concentration from 0 to 0.075% (v/v) and subsequently the values decreased beyond 0.075% (v/v) soybean oil (Table 1). In the present system, the maximum increase in  $K_L a$  with respect to zero soybean oil concentration was 1.3 fold. The phenomenon of increase in  $K_L a$  up to certain

concentration of soybean oil followed by a decrease was suspected to be dependent on the distribution of NAPL in the present system based on visual observation. At soybean oil below 0.075% (v/v), most of the soybean oil existed on the surface of the beaker, whereas beyond 0.075% (v/v), free soybean oil droplets were observed on the water surface. This change in distribution of soybean oil beyond 0.075% (v/v) may cause lowering of the  $K_{L,a}$ . This can be explained with the help of spreading coefficient as explained in section 1. However, spreading coefficient determination was not carried out in the present study. Similar observations were reported in literature when soybean oil was used as oxygen vector by other researchers. At low concentration,  $K_{L,a}$  was found to be proportional to the volumetric oil fraction, whereas, at high concentration,  $K_{L,a}$  reached a maximum value and then decreased slightly [2]. The  $K_{L,a}$  value was found to be maximum at 2% soybean oil and decreased significantly beyond this concentration during production of tetracycline in a air-lift bioreactor [8]. Similar observations were reported when different types of hydrocarbons were used as oxygen vectors by various researchers. When a mixture of n-alkanes was added as oxygen vector in a tower bioreactor, 4% liquid hydrocarbons resulted in maximum  $K_{L,a}$  and subsequently decreased due to accumulation of liquid hydrocarbons in the liquid phase [3]. An increase in n-C16 beyond 2% (v/v) volume did not show further improvement on penicillin fermentation [6]. Decrease in oxygen transfer rate was observed for n-C16 concentration beyond 0.27% in a bioreactor with six-blade Rushton turbine impellers [9]. However, no explicit reason was reported for this phenomenon.

Subsequently, OUR, also known as the oxygen consumption or respiration rate, which is defined as the milligram of oxygen consumed per gram of volatile suspended solids (VSS) per hour was measured (Fig.3). OUR in the presence of 0.1% soybean oil (v/v) was found to be 0.006853 mg O<sub>2</sub>/mg MLSS/hr compared to the control system with 0.004795 mg O<sub>2</sub>/mg MLSS/hr. Increased oxygen uptake rate in the presence of oil shows increased metabolic activity of MLSS in the presence of soybean oil.

This is the first study reporting the effect of soybean oil on oxygen mass transfer phenomenon in wastewater treatment. The increase in  $K_{L,a}$  in the presence of soybean oil may result in reducing the energy requirement in conventional biological treatment systems. However, the effect of film formation versus droplet formation on oxygen mass transfer in such system and its impact on wastewater treatment is not fully elucidated and more research is required to understand this phenomenon.

Table 1. Variation in  $C_s$  and  $K_{L,a}$  at various soybean oil concentration based on three-parameter non-linear regression

Soybean oil concentration (%, v/v)	Temp. (°C)	$C_o$ (mg/L)		$C_s$ (mg/L)	MLSS (mg/L)	$K_{L,a}$ (min <sup>-1</sup> )
		Expt.	Fitted	Fitted (mg/L)		
0	24	0.12	0.35	7.62	160.3	0.007
0.01	24	0	0	8.52	150.1	0.0071
0.03	24	0.41	0.39	7.84	178.8	0.008
0.05	22	0.14	0.18	7.62	184.6	0.0078
0.075	22	0.09	0.19	7.68	182.8	0.0091
0.1	24	0.23	0.46	8.60	171.4	0.0064
0.3	23	0	0	8.33	172.7	0.0084
0.5	24	0	0	7.84	188.8	0.0078
0.75	24	0	0	8.13	188.8	0.0055

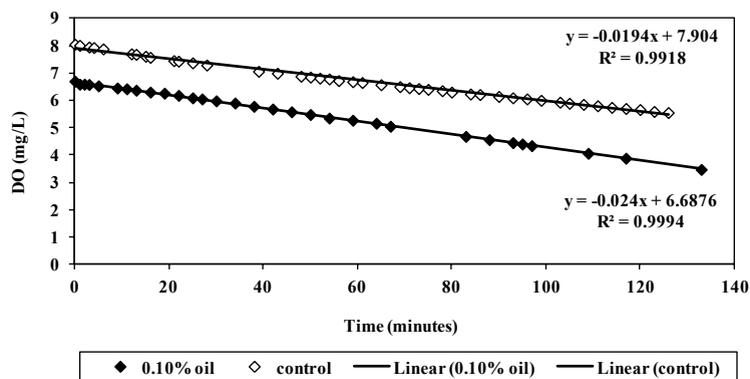


Fig. 3. Oxygen uptake rate in the absence and presence of 0.1% soybean oil

#### 4. Conclusion

Optimum paddle speed and MLSS concentration for carrying out oxygen transfer experiment in the presence of soybean oil was found to be 75 rpm and 150-200 mg/L. Three parameter non-linear regressions were found to provide better fit compared to linear regression. Maximum  $K_La$  has been observed when soybean oil concentration was 0.075% (v/v).  $K_La$  was found to increase by 30% in the presence of soybean oil compared to absence of soybean oil.  $K_La$  increased with increase in soybean oil concentration up to a certain point (0.075% v/v) and was found to decrease subsequently.

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