Effects of carrier-attached biofilm on oxygen transfer efficiency in a moving bed biofilm reactor

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Abstract Three laboratory-scale moving bed biofilm reactors (MBBR) with different carrier filling ratios ranging from 40% to 60% were used to study the effects of carrier-attached biofilm on oxygen transfer efficiency. In this study, we evaluated the performance of three MBBRs in degrading chemical oxygen demand and ammonia. The three reactors removed more than 95% of NH_4^+ -N at an air flow-rate of $60 \text{ L} \cdot \text{h}^{-1}$. The standard oxygen transfer efficiency (αSOTE) of the three reactors was also investigated at air flow-rates ranging from 60 to $100 \mathrm{L} \cdot \mathrm{h}^{-1}$. These results were compared to α SOTE of wastewater with a clean carrier (no biofilm attached). Results showed that under these process conditions, αSOTE decreased by approximately 70% as compared to αSOTE of wastewater at a different carrier-filling ratio. This indicated that the biofilm attached to the carrier had a negative effect on αSOTE. Mechanism analysis showed that the main inhibiting effects were related to biofilm flocculants and soluble microbial product (SMP). Biofilm flocs could decrease αSOTE by about 20%, and SMP could decrease αSOTE by 30%–50%.

Keywords carrier, biofilm, oxygen transfer efficiency, moving bed biofilm reactor

1 Introduction

Due to increased flow and organic loading, many wastewater treatment plants are faced with facility upgrades and retrofits to provide additional capacity [\[1](#page-7-0)]. At the same time, the moving bed biofilm reactor (MBBR) is receiving increased attention due to its provision of a carrier with a high specific surface, for formation of biofilm, and for a more flexible and compact reactor [\[2\]](#page-7-0). The primary

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advantage of MBBRs over activated sludge reactors is their compactness and elimination of the need for sludge recirculation [\[3](#page-7-0)]. The MBBR is more resistant to organic shock and can improve the efficiency of $NH₄⁺ - N$ removal [[4,5\]](#page-7-0). The aeration unit is important for wastewater treatment in MBBRs. It not only provides enough dissolved oxygen (DO) for microorganisms, but also stirs the mixture evenly. However, aeration also constitutes the biggest energy-consuming step in the MBBR process, which accounts for $45\% - 75\%$ of the total energy consumption [[6](#page-7-0)–[8](#page-7-0)]. Thus, the aeration efficiency, which depends on the oxygen transfer efficiency (OTE) in the aerated reactors, directly affects the cost of the wastewater treatment process. The main factors that affect OTE in wastewater treatment are reactor design (i.e., depth and shape), characteristics of the wastewater (i.e., suspended solids, temperature, and viscosity), aeration system (i.e., distribution of aeration system and type of diffusers), and ambient conditions (i.e., altitude and temperature) [[9](#page-7-0)–[12\]](#page-7-0). Suspended carrier elements can also affect OTE during the MBBR process.

A few publications have provided evaluations of the effect of carrier elements on OTE in clean water with varying results, indicating that addition of carriers affects OTE. Pham et al. [[13](#page-7-0)] demonstrated that increasing the filling ratio of carriers decreased OTE, while Jing et al. [[14](#page-7-0)] showed that the oxygen transfer coefficient (K_La) increased with an increase in the carrier-filling ratio. After studying a full-scale integrated fixed film activated sludge system (IFAS), Viswanathan et al. [[15](#page-7-0)] concluded that the carriers had little or no effect on OTE in fine bubble systems. Until now, little information has been available on the impact of carrier elements on OTE in the MBBR process, and none of the studies compared OTE under wastewater conditions and in MBBRs. In addition to the carriers, the biofilm attached to the carriers could also significantly influence OTE. However, there is no published work on the influence and mechanism of biofilm effects on OTE in MBBRs.

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Therefore, the two main objectives of this study were to investigate the effect of carrier-attached biofilm on OTE in the MBBR at different carrier filling ratios (from 40% to 60%), and at different air flow-rates (from 60 to $100 \mathrm{L} \cdot \mathrm{h}^{-1}$). In this study, we tried to demonstrate the influence of biofilm on OTE by comparing the standard oxygen transfer efficiency $(\alpha$ SOTE) under wastewater conditions with the αSOTE under process conditions. The mechanism by which biofilm influences OTE was also investigated.

2 Materials and methods

2.1 Set-up and operation of the MBBR

The laboratory scale, MBBR system used to cultivate the biofilm carriers throughout this study is shown in Fig. 1. Each MBBR reactor was a Plexiglass cylinder with effective volume of 8.66 L and internal diameter of 17 cm. Suspended carriers were made of polyethylene with a density close to $1 \text{ g} \cdot \text{cm}^{-3}$. The plastic carrier was cylindrical (length 25 mm and diameter 10 mm) with a cross inside the cylinder and fins outside. To provide oxygen to the water and fluidize the biofilm carriers, an aerator was fixed at the center bottom of the reactor. The air flow-rate was monitored by a calibrated rotameter. The dissolved oxygen concentration (DO) was measured using an oxygen electrode (WTW, German) that was calibrated daily. In the experiments, three sets of MBBRs were operated in parallel with different carrier-filling ratios

(40%, 50%, and 60%). The hydraulic retention time (HRT) of all reactors was 7 h. The temperature was kept at 26°C by a water heater (Visi-Therm, USA).

Three reactors were inoculated with activated sludge from the Beixiaohe Wastewater Treatment Plant in Beijing. Initially, the carriers and inoculating activated sludge were both placed in each reactor. The sludge was allowed to settle for 24 h to provide enough contact between the carrier and inoculating microorganisms, and then was removed. All reactors were operated as continuous flow systems with small influent flows. When the systems were stable, the influent was gradually increased. All reactors were fed synthetic wastewater to maintain COD between 300 and 350 mg· L^{-1} and ammonium concentration in the influent at approximately $30 \text{ mg} \cdot L^{-1}$. The other nutrient concentrations were as follows (per $55 L$): CaCl₂ (1.5 g); $Mg \cdot 7H_2O$ (3 g); NaHCO₃ (7.9 g). Trace elements were simultaneously added to the influent to facilitate the growth of microorganisms [\[16\]](#page-7-0). The pH in the reactor ranged from 7.2 to 7.5 during the experiments. The biofilm was cultivated on the carrier after approximately 90 days.

The experiments reported below were carried out only after the MBBR parameters indicated excellent and stable performance. Table 1 shows the experimental processes.

2.2 Analytical methods

2.2.1 Water quality

During continuous operation of the MBBRs, effluent

Fig. 1 (a) Schematic of laboratory MBBR system and off-gas analysis equipment; (b) suspended carrier

Table 1 Experimental conditions applied in all tests

| tests | aeration rate $/(L \cdot h^{-1})$ | batch reactors | | | | |
|----------------------------|--------------------------------------|----------------|-----|-----|-------------|--|
| | | | | | time $/d$ | aim |
| different filling ratio | 60 | 40% | 50% | 60% | $190 - 211$ | to investigate the effect of carrier-attached biofilm with different filling ratio on OTE |
| different air flow rate | 60 | | 50% | | 190–211 | to investigate the effect of carrier-attached biofilm with different air flow rate on OTE |
| | 80 | | 50% | | $214 - 225$ | |
| | 100 | | 50% | | $228 - 240$ | |

samples were immediately filtered through 0.45 μm filter paper and then analyzed. Soluble COD and NH_4^+ -N were measured according to the Standard Method for Water and Wastewater Examination [\[17\]](#page-7-0). The total suspended solids of biofilm biomass (MLSS) were measured using the method reported by Li et al. [\[5](#page-7-0)]. Five carriers were removed from, and five other carriers added to, the MBBRs in order to maintain a constant filling ratio in the reactor. The oxygen uptake rate (OUR) was used to represent biofilm activity, and it was measured using the methods reported by Joanna et al. [[18](#page-7-0)]. For each measurement, five carriers and 250 mL of the mixture were used. The specific oxygen uptake rate (SOUR) was defined as the milligrams of oxygen consumed per gram of suspended solids (SS) per hour in the test.

2.2.2 Soluble microbial product (SMP) analysis

Briefly, the untreated, mixed liquid samples from each reactor were centrifuged to represent SMP [\[19\]](#page-7-0). Carbohydrate concentrations in the processed samples were quantified using the anthrone method, with dextrose used as a standard [[20](#page-7-0)]. The proteins were determined according to a modification of the Lowry method [\[21](#page-7-0)].

2.2.3 OTE analysis

The oxygen transfer efficiency (OTE) was monitored using the off-gas analyzer. It was derived from the off-gas method employed to determine oxygen transfer in the activated sludge process [[22](#page-8-0)]. OTE was calculated using the following equations:

$$
OTE = \frac{mass\ O_2\ in-mass\ O_2\ out}{mass\ O_2\ in} = \frac{G\cdot M_{Ri} - G\cdot M_{Re}}{G\cdot M_{Ri}},\quad (1)
$$

$$
OTE = \frac{M_{Ri} - M_{Re}}{M_{Ri}},\tag{2}
$$

where G, M_{Ri} , and M_{Re} were the mass of nitrogen and inert gas, the molar fractions of oxygen in the inlet gas, and the molar fractions of oxygen in the off gas, respectively.

$$
M_{Ri} = \frac{Yi}{1 - Y_i - Y_{Ci} - Y_{Wi}},
$$
\n(3)

$$
M_{Re} = \frac{Y_e}{1 - Y_e - Y_{Ce} - Y_{We}},\tag{4}
$$

where Y_i and Y_e are the molar fractions of O_2 in the inlet gas and off gas; Y_{Ci} and Y_{Ce} are the molar fractions of $CO₂$ in the inlet gas and off gas; and Y_{W_i} and Y_{W_e} the molar fractions of water in the inlet gas and off gas.

As the molar fraction of oxygen in the supplied air is known ($Y_i = 0.2095$), the OTE can be determined by

measurements of the oxygen molar fraction in the off gas, as well as the $CO₂$ and water molar fraction in the inlet gas and off gas, respectively.

To make comparison of different conditions feasible, the measured OTE was converted to standard oxygen transfer efficiency $(\alpha$ SOTE), and calculated using the following equation:

$$
\alpha SOTE = OTE \times \frac{C_{\infty,20}^*}{C_{\infty,t}^* - C_L} \times \theta^{(20-T)},\tag{5}
$$

where α SOTE is the standard oxygen transfer efficiency under test; C_L the oxygen concentration during testing; $C_{\infty,t}$ the oxygen concentration in equilibrium with the supplied air at test conditions; and $C_{\infty,20}$ the oxygen concentration at saturation, expressed at standard conditions.

In this study, the αSOTE measured in MBBRs with carrier-attached biofilm was called αSOTE under process conditions, and the αSOTE of wastewater with clean carriers was called αSOTE under wastewater conditions. The difference between the two conditions was whether there was biofilm attached to the carriers. With regard to the process conditions, artificial wastewater, which contained a variety of nutrients and trace elements, was used as the influent of the MBBRs. To strictly control variables and eliminate the effect of water quality, tests under wastewater conditions were conducted after adding clean carriers to wastewater.

3 Results and discussion

3.1 MBBR performance

Figure 2 shows variations of COD and $NH₄⁺ -N$ in the influent and effluent of three reactors. As illustrated in Fig. 2, COD removal efficiency was about 70%, whereas NH_4^+ -N removal efficiency was more than 95%. In this experiment, the treatment efficiency of NH_4^+ -N was excellent regardless of carrier filling ratios, in agreement with the findings of Duan et al. [\[19\]](#page-7-0). Without discharging sludge, microorganisms gradually accumulated on the surface of carriers and formed biofilm, which was beneficial to the growth of nitrifying bacteria [[23\]](#page-8-0). The results indicated that the MBBR process efficiently removes NH_4^+ -N.

3.2 Effect of carrier-attached biofilm on oxygen transfer efficiency

In MBBRs, both the carrier filling-ratio and air flow-rate were important operating conditions. The air flow-rate, in particular, was the main factor affecting oxygen transfer efficiency [[24](#page-8-0)]. Therefore, we investigated the effect of

Fig. 2 Variation of COD and NH $_4^+$ -N in the influent and effluent, and removal efficiency at three different carrier filling ratios: (a), (b) 40% carrier filling ratio; (c), (d) 50% carrier filling ratio; (e), (f) 60% carrier filling ratio

carrier-attached biofilm on OTE in the MBBRs at different carrier filling-ratios, ranging from 40% to 60%, and at different air flow-rates, ranging from 60 to $100 \mathrm{L} \cdot \mathrm{h}^{-1}$.

The effect of clean carriers without attached biofilm on OTE in clean water had already been investigated. Viswanathan's study showed that clean-carrier filling ratios ranging from 20% to 60% had no significant effect on OTE in clean water[\[15\]](#page-7-0).

Researchers found that high biomass concentration affected OTE in MBRs (i.e., by reducing oxygen transfer) [[25](#page-8-0)]. Similarly, the biofilm attached to carriers may affect OTE due to the presence of biofilm flocs. Therefore, in this

study, we also evaluated the effect of biomass concentration on OTE.

3.2.1 Variations in carrier filling ratio

Figure 3 shows the oxygen transfer efficiency of the aeration system at three different carrier filling ratios. Results showed that the α SOTE under process conditions were clearly lower than that under wastewater conditions. Compared to αSOTE under wastewater conditions, the αSOTE under process conditions decreased approximately 70%. This indicated that the biofilm attached to carriers negatively affected OTE.

Until now, there has been little research on the effect of biofilm on OTE. Many researchers had investigated OTE in activated sludge systems, but little information was available about OTE under process conditions in MBBRs. In activated sludge systems, the main factor influencing OTE involved surface-active agents [[26](#page-8-0)]. In contrast, there were no surfactants in the influent of the MBBRs. This implied that the biofilm itself influenced the OTE in MBBRs.

The αSOTE under process conditions was lower than αSOTE under wastewater conditions because the biofilm attached to the carrier decreased OTE in the MBBR. It was concluded that the air flow-rate in MBBRs might be higher than that in activated sludge systems. Moreover, because it was more difficult for the dissolved oxygen to pass through a certain thickness of biofilm for microbial respiration in an MBBR, the air flow-rate should be higher to maintain a higher DO concentration.

In addition, there was no significant change observed in the αSOTE values under process conditions at different filling ratios. This might also demonstrate that the carrier itself had no effect on OTE.

3.2.2 Variations in air flow rate

The αSOTE under wastewater and process conditions, at 50% carrier filling ratio, and with different air flow-rates are shown in Fig. 4. As the air flow-rate increased from 60 to $100 \mathrm{L} \cdot \mathrm{h}^{-1}$, the α SOTE under wastewater conditions decreased. However, under process conditions, the change in α SOTE (~ 2%) was insignificant. Compared to wastewater conditions, whatever the air flow-rate in the tests, the αSOTE under process conditions was noticeably less. This also indicated that the biofilm attached to carriers had a negative effect on OTE.

Under wastewater or clear water conditions, the air flowrate was one of the most important factors influencing OTE, and αSOTE decreased with increasing rate of air flow [10[,24\]](#page-8-0). However, under process conditions, microbial respiration as well as the air flow-rate, could affect OTE. Anand [\[27\]](#page-8-0) reported that cell respiration enhanced the oxygen transfer rates. As shown in Fig. 5, there was a

Fig. 3 Oxygen transfer efficiency at three different filling ratios under process and wastewater conditions: (a) αSOTE of 40% reactor, (b) αSOTE of 50% reactor, (c) αSOTE of 60% reactor; αSOTE under wastewater conditions (Δ), and αSOTE under process conditions (\blacksquare) of each reactor is shown in (a–c)

better linear relationship between αSOTE under process conditions and OUR ($R^2 = 0.96$). Generally, α SOTE increased with increasing OUR. When OUR increased from around $15 \text{ mg} \cdot (\text{L} \cdot \text{h})^{-1}$ to around $25 \text{ mg} \cdot (\text{L} \cdot \text{h})^{-1}$, αSOTE increased from 2.0% to 3.2%. This indicated that microbial respiration could increase OTE.

In this study, OUR was measured in real time and the

Fig. 4 α SOTE under wastewater and process conditions with different air flow-rates: α SOTE under wastewater conditions (Δ); α SOTE under process conditions (\blacksquare)

biofilm biomass was stable. Therefore, the measured OUR represented microbial respiration, which could increase OTE. In addition, the density of the carrier was higher after attachment of biofilm, which promoted fluidization, and at a high rate of air flow, strong turbulence was more conducive to mass transfer. These two aspects caused an insignificant reduction in αSOTE under process conditions.

However, Hu [\[28\]](#page-8-0) concluded that the α-value (α SOTE/ SOTE) decreased as the OUR of the activated sludge increased in an MBR. Hu's observation could be interpreted as an effect of MLSS concentration on oxygen transfer. In the MBR, the OUR of the activated sludge increased as the MLSS concentration increased. As discussed previously, the OTE in our experiments decreased with higher MLSS concentration. Therefore,

the effect of OUR on the α-value was actually the effect of MLSS.

3.3 Interpretation of biofilm on oxygen transfer efficiency

According to test results using different filling ratios and air flow-rates, the αSOTE under process conditions was significantly lower than that under wastewater conditions. The results showed that biofilm attached to the carrier had an adverse effect on oxygen transfer efficiency. The effect of biofilm on the oxygen transfer efficiency could be generalized as the influence of biofilm biomass, biofilm flocs, microbial respiration, SMP secreted by biofilm, and the aging suspended products of biofilm flocs. Among these, the aging suspended flocs were negligible. It is important to note that microbial respiration could also increase oxygen transfer efficiency. Therefore, the study only considered the most probable main inhibitory factors: biomass concentration, biofilm flocs, and SMP.

3.3.1 Effect of biomass concentration on oxygen transfer efficiency

The growth in the biofilm biomass of the MBBR with 40% carrier filling-ratio and $40 \mathrm{L} \cdot \mathrm{h}^{-1}$ air flow-rate is shown in Fig. 6(a). As shown in Fig. 6(b), α SOTE under process conditions changed insignificantly with increase in biofilm biomass, which could be related to microbial respiration. When biofilm biomass was high, microbial respiration required more oxygen. Hence, αSOTE could remain high for a high OUR. While biofilm was growing, biomass lessened. In this case, biofilm activity could also be elevated because of high substrate loading. It has been determined that biofilms grown under high substrate loading have significantly higher activity [[29](#page-8-0)]. Less biomass means more substrate loading. Consequently, SOUR may have been high as biofilm was growing, which can be verified in Fig. 6(c). Therefore, there was no direct correlation between αSOTE under process conditions and biofilm biomass.

3.3.2 Effect of biofilm flocs on oxygen transfer efficiency

To avoid the effect of microbial respiration on oxygen transfer efficiency, biofilm (with the carrier it was on) was taken from the reactor, and then placed in clean water to measure its OTE. The αSOTE of carrier-attached biofilm in clean water was initially 4.97%, whereas the α SOTE of the same number of clean carriers without biofilm was 6.12%. Because biofilm flocs attached to the carrier hindered the movement of air bubbles (compared to a carrier without biofilm), the biofilm flocs could reduce OTE by about 20%. Biofilm was growing on the surface of the carrier, and after a certain time, the biofilm thickness increased. After air bubbles were released from the aerator, they rose

Fig. 6 (a) Biofilm biomass of 40% carrier filling ratio during tests; (b) the correlation between αSOTE under process conditions and biofilm biomass; (c) the correlation between OUR and MLVSS

and might be held up when moving past the biofilm flocs. The air bubbles could not move smoothly past the inside surface of the carrier, which held the bubbles back and led to coalescence that formed larger bubbles, thereby lowering the oxygen transfer efficiency.

3.3.3 Effect of SMP on oxygen transfer efficiency

Figure 7(a) shows that α SOTE under wastewater, process, and effluent conditions were 6.51%, 1.73%, and 3.76%, respectively. Compared to αSOTE under wastewater conditions, αSOTE under effluent conditions was reduced by 42%. This confirmed that some substance in liquid phase decreased oxygen mass transfer efficiency under process conditions. This substance might be the soluble microbial product (SMP) secreted by biofilm. The SMP is a pool of organic compounds released into solution from substrate metabolism, usually related to growth and decay of microbial biomass [\[30](#page-8-0)]. SMP was primarily composed of proteins, humic compounds, and polysaccharides. To reach the active sites of bacterial cell membranes, the oxygen contained in the air bubbles needed to penetrate the liquid film surrounding the SMP flocs [\[31\]](#page-8-0). Therefore, SMP was likely to affect oxygen transfer efficiency. Figure 7(b) shows the effect of SMP on OTE. This indicated that the effect of SMP on αSOTE was related to the concentration of SMP. When the concentration of SMP was lower than $25 \text{ mg} \cdot L^{-1}$, the inhibitory effect was less significant and αSOTE decreased by about 30%. In contrast, when the concentration of SMP exceeded 25 mg \cdot L⁻¹, the inhibitory effect was stronger and α SOTE decreased by about 50%.

4 Conclusions

In this study, we investigated the effects of carrier-attached biofilm on oxygen transfer efficiency. Results showed that αSOTE under process conditions was significantly lower than under wastewater conditions at different filling ratios and air flow-rates. This indicated that the biofilm attached to carriers had a negative effect on oxygen transfer efficiency, and decreased αSOTE about 70% (from about 7.0% under wastewater conditions, to about 2.0% under process conditions). Mechanism analysis showed the main inhibiting effects were related to biofilm flocs and SMP.

Fig. 7 (a) αSOTE under process, wastewater, and effluent conditions; (b) the effect of SMP on αSOTE

Biofilm could decrease αSOTE by about 20%, and SMP by 30%–50%. In addition, microbial respiration could increase oxygen transfer efficiency. There was a better linear relationship between αSOTE under process conditions and OUR ($R^2 = 0.96$).

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