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Investigation of membrane-aerated biofilm reactor (MABR) for the treatment of crude oil wastewater from offshore oil platforms

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ABSTRACT

Membrane-aerated biofilm reactor (MABR) was designed to treat oilfield produced wastewater from offshore oil platform. The effects of suspended solids (SS) of influent, intra-membrane air pressure, and feed flow velocity on chemical oxygen demand (COD), oil, and ammonia nitrogen (NH_4^+-N) removals were investigated through a 160-d process study. The results indicate: (a) high influent SS resulted in the formation of a barrier layer containing insoluble SS on the outside surface of biofilm which significantly reduced the mass transfer of NH_4^+-N and organic substrates from wastewater to the biofilm; (b) under the condition of feed flow velocity of 0.01 m/s, with the increase in intra-membrane air pressure, the oxygen permeate flux of hollow fiber membrane was enhanced and NH_4^+-N removal efficiency at 0.12 MPa was up to 95%; (c) under the condition of intra-membrane air pressure of 0.08 MPa, with the increase in flow velocity, the performance of MABR was significantly enhanced, the best removal efficiencies of COD, oil, and NH_4^+-N reached 60.3, 80, and 95%, respectively, at the feed flow velocity of 0.05 m/s.

Keywords: MABR; Intra-membrane air pressure; Crude oil wastewater; Feed flow velocity

1. Introduction

In recent years, various technologies including polymer (mainly hydrolyzed polyacrylamide (HPAM)) flooding have been extensively used in China to enhance oil recovery. However, no matter which way we choose to extract oil, a large amount of oil field wastewater was produced in the oil production process. A large part of the wastewater was injected back into the stratum for reuse [1] and the rest was discharged into the environment after simple treatment, which had negative effects on the water environment.

Oil field produced wastewater from offshore oil (OPWFOOP) is very complex, which contains high levels of oil, salinity, residual HPAM, suspended solids (SS), sulfide, and reductive substances, etc. The remained HPAM in the wastewater not only enhances the stability of oil–water emulsion, but also contributes to the total chemical oxygen demand (COD) of the wastewater. The high salinity makes the wastewater

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more difficult to be treated. Currently conventional wastewater treatment techniques cannot deal with this kind of wastewater effectively, so a new method to treat OPWFOOP should be developed.

Membrane bioreactor was proved feasible for treating surfactant containing oil water emulsions [2]. Flocculation and ceramic membrane filtration were used for the treatment of oily wastewater [3]. A novel cross flow oil–water separator was proposed to treat oil produced water [4]. In recent years, Membrane-aerated biofilm reactor (MABR) has been widely studied to deal with a variety of wastewater, such as removal of aromatics [5], phenol [6] and acetonitrile [7], treatment of hyper-saline wastewater [8], slightly polluted surface water [9], and high-strength industrial sewage [10]. However, reports about the study of MABR to deal with OPWFOOP are rare.

The difference between MABRs and conventional biofilm reactors is highlighted by the concept of active layers within biofilms and the importance of active layer location [11]. The main advantage of MABRs over conventional biofilm reactors is that oxygen diffusing through the gas permeable membrane without any bubble formation, which is favorable for the growth of biofilm attached to the membrane's exterior. Moreover, membrane aeration allows changing the amount of oxygen supply by adjusting air pressure or flow rate, which reinforces rigorous control of the system [12,13].

The MABR has three zones: bulk wastewater, biofilm, and membrane. In the biofilm process, an oxygen concentration gradient is created across aggregated micro-organism, so that both aerobic and anaerobic conditions can be established inside the biofilm, and where the aerobic and anaerobic layer are located adjacent to the biofilm–membrane interface and the biofilm–liquid interface, respectively [14]. The removal of ammonia nitrogen (NH_4^+-N) was carried out by being converted into nitrate or nitrite during nitrification, and then converted into nitrogen during denitrification [15,16]. Simultaneous nitrification and denitrification in a single reactor using a MABR was proposed [17]. The intra-membrane oxygen pressure was found to be one of the two most important parameters influencing the MABR performance [18].

Organic substances in wastewater, such as aromatic hydrocarbons, chlorinated organics, hydrocarbons, nitrogen compounds, and other contaminants, must be transferred from the bulk liquid into the interior of biofilm and then be degraded or absorbed by micro-organisms in biofilm [19,20]. Flow velocity influenced mass transfer in the diffusion boundary layer and it was one of the key factors affecting the performance of MABR [21].

In this study, we attempt to evaluate the feasibility of MABR for the treatment of OPWFOOP. A cross flow of MABR system was designed. The removal rates and efficiencies of COD, oil, and NH_4^+-N were investigated experimentally under different operating conditions.

2. Materials and methods

2.1. Materials

The hollow fiber membranes with average pore size of 0.2 μm were provided by Tianjin Hydroking S&T Ltd. (Tianjin, China). Engineered bacteria AD350M (Advance Biotechnologies company, Canada) and long-sludge-age activated sludge (Tianjin University, China) were used to form biofilm on the outside surface of hollow fiber membrane. The wastewater was taken from one oil platform on the Bohai Sea.

2.2. Experiment of biofilm growth

One gram of engineered bacteria AD350M was activated through three-day's continuous aeration in a beaker containing deionized water. Then, the activated engineering bacteria and the activated sludge (Tianjin University, China) in a volume ratio of 1:1 were added into the MABR. The feed flow velocity was 0.01 m/s. Another 3 d later, mixed liquid with different ratio of nutrient solution and OPWFOOP which was pretreated with poly aluminum chloride (PAC) and polyacrylamide (PAM). Ten percent PAC solution was added into OPWFOOP by a ratio of 2 ml/L (v/v, PAC solution/ OPWFOOP) with rigorous stirring. Then, 1‰ PAM solution mixed with that solution by a ratio of 0.5 ml/L (v/v, PAM solution/OPWFOOP). PAC

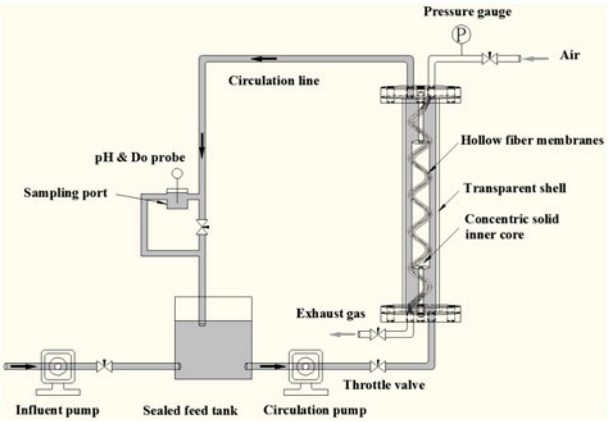

Fig. 1. Schematic diagram of the MABR used in this study.

Table 1
Operation conditions of the bioreactor during the process study of MABR

Condition	Days of operation (d)	Influent NH_4^+-N (mg/L)	Influent COD (mg/L)	Influent oil (mg/L)	SS (mg/L)	Intra-membrane air pressure (MPa)	Intra-membrane air flow rate (m/s)	Feed flow velocity (m/s)	HRT (h)
1	30–44	30.8	380–450	24–30	40–80	0.1	0.0108	0.01	24
2	45–53	–	–		–	–		–	–
3	54–74	28.2	276–295	9–17	12–18	0.02	0.0022	0.01	24
4	75–93	29.1	297–312	11–19	15–21	0.08	0.0072	0.01	24
5	94–116	30.8	278–311	10–19	12–16	0.12	0.0145	0.01	24
6	117–140	30.0	287–314	9–18	12–17	0.08	0.0072	0.02	24
7	141–160	30.3	278–320	9–21	9–17	0.08	0.0072	0.05	24

and PAM worked as flocculants to remove colloid, oil particles, and SS from the OPWFOOP. And, the ratio of OPWFOOP/nutrient solution varied from 1:10, 1:5, 1:4, 1:3, 1:1, 2:1, 3:1, 4:1 to 1:0. The circulation times of different mixed liquid in the MABR were 3, 2, 2, 2, 3, 3, 2, 3, and 3 d, respectively. After 29 d of operation, the formation of biofilm on membrane surfaces was accomplished and biofilm adapted to the saline environment. The growth process of biofilm was observed by visual observations.

2.3. Experiment of MABR

The MABR system shown in Fig. 1 was composed of a membrane module, a cylindrical plastic shell with the length of 500 mm, and the inner diameter of 50 mm. The membrane module was made by winding 100 hollow fiber membranes with the length of 1.2 m on a plastic core bar with the diameter of 30 mm. The two ends of the hollow fiber surfaces were sealed in two short plastic tubes with epoxy resin, respectively. Then, solid core bar with the hollow fibers were assembled in the module shell by fixing the fiber sealing tubes and plastic core bar to the module shell headers with epoxy resin, respectively. The hollow fiber membranes were loose and dispersed in the annular channel. Compressed air was supplied by connecting one end of the hollow fiber membrane with an air compressor. A throttle valve was installed on another end of hollow fiber membrane to ensure an appropriate gas flux and pressure. The relation between air flow rate and intra-membrane pressure is shown in Table 1. The wastewater was pumped into the bottom of bioreactor from a reservoir, and then circulated in the MABR system. The flow direction is almost perpendicular with the hollow fiber membranes.

In order to get better growth of bacteria and better removal efficiencies, some nutrients added into the wastewater was necessary, these compounds were as follows: KH_2PO_4 26 mg/L, K_2SO_4 45 mg/L, $MgSO_4 \cdot 7H_2O$ 55 mg/L, $NaHCO_3$ 300 mg/L, Na_2CO_3 100 mg/L, Fe_2 (SO4)$_3$ 0.1 mg/L, $CuSO_4 \cdot 5H_2O$ 0.1 mg/L, $MnSO_4$ 0.1 mg/L, and $ZnSO_4$ 0.1 mg/L. During the MABR process study, the influent temperature was maintained at a room temperature of 23–25℃.

OPWFOOP circulation was carried out by a peristaltic pump (BT01-YZ2515, Tianjin Xieda Electronic Co., Ltd., China). Oil concentration was measured by an infrared photometric oil in water analyzer (Jky-2A, Jilin Jike Instrument Co., Ltd., China). Ammonia nitrogen (NH_4^+-N), nitrate and nitrite concentrations were measured by spectrophotometer using multi-parameter iron concentration measuring device (HI83200, Hanna Instruments Inc., USA). Dissolved oxygen (DO) concentration was measured by a portable DO meter (FG4-ELK, Mettler-Toledo Instruments (Shanghai) Co., Ltd., and China). COD concentration was measured by a COD multifunction measuring device (ET3150B+ET1150, Eurotec Ltd., China).

3. Results and discussion

Table 1 shows the operation conditions of the MABR during seven different test periods. The performances of the bioreactor were monitored by measuring the concentrations of COD, oil, and NH_4^+-N.

As shown in Fig. 2(a) 160 d process study was carried out to investigate the effect of high SS, intra-membrane air pressure, and flow velocity on COD, oil, and NH_4^+-N under different operation conditions and hydraulic retention time of 24 h. Fig. 2(a) shows time course for COD and NH_4^+-N concentrations in the MABR. In this study, insoluble oil was removed by adding flocculants to form agglomerates which were removed by precipitation in the pretreatment state. The soluble oil circulating in MABR system attached on the surface of biofilm. Bacteria lived in biofilm and

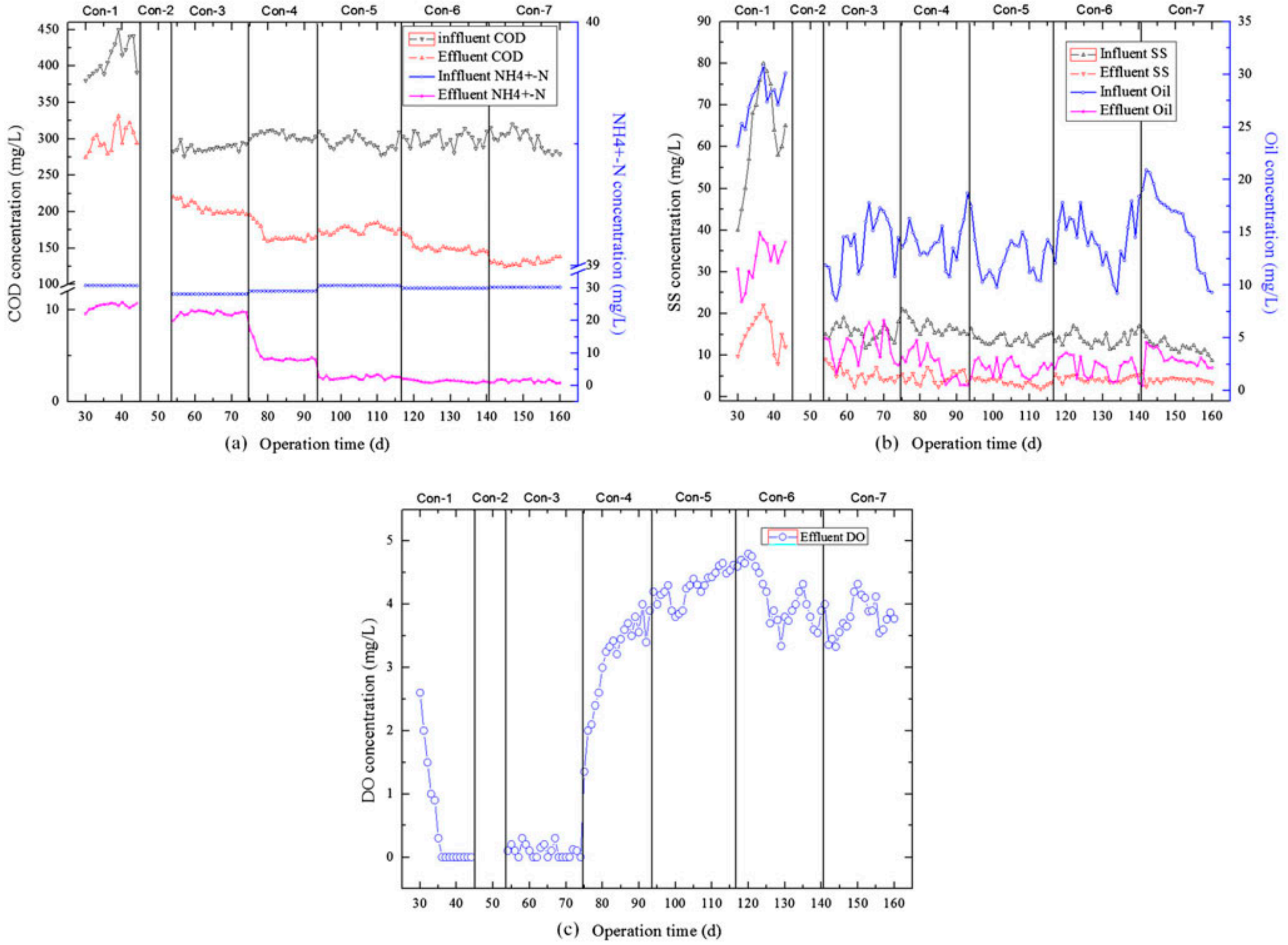

Fig. 2. Variations of COD, NH_4^+-N, SS, and oil with operation time at different operation conditions. (a) COD and NH_4^+-N concentrations in the MABR influent and effluent; (b) SS and oil concentrations in the MABR; (c) DO concentration in the MABR effluent.

degraded these oil into H_2O and CO_2 eventually. During **Con-1**, the average removal rates of COD and NH_4^+-N were just 33.3 and 20.4%, respectively, because organic substrates and NH_4^+-N cannot be effectively diffused from bulk to the biofilm. However, the average removal rates of oil and SS reached to 60.2 and 80.6% (see Fig. 2(b)) for oil and SS almost adsorbed on the outside surface of the biofilm and formed a barrier layer (BL). The effluent DO concentration was lower than 0.5 mg/L (see Fig. 2(c)). The influent used in the following experiments was pretreated by flocculation.

In order to investigate the effects of intra-membrane air pressure on the performance of the MABR, aeration pressure experiments were carried out (**Con 3–5**) at the feed flow velocity of 0.01 m/s, with substantially no change in the quality of influent and other operating conditions. Before the aeration pressure experiments, a nine-day experiment (**Con-2**) for detaching the BL from outside surface of biofilm and recovering the performance of reactor was run. During **Con-3**, effluent NH_4^+-N was above 20 mg/L and the average removal efficiency was just 21.0%. Effluent COD was above 195 mg/L and the average removal efficiency was just 28.7%.

The intra-membrane air pressure was increased from 0.02 MPa to 0.08 and 0.12 MPa in **Con-4** and **Con-5**, respectively. Fig. 2(a) shows that the average removal rates of NH_4^+-N and COD were significantly enhanced with the increase in intra-membrane pressure. Effluent NH_4^+-N were reduced to 8 and 3 mg/L and the average removal efficiencies were increased to 68.8 and 92.0%, respectively. Effluent COD were reduced to 165 and 175 mg/L and the average removal efficiencies were 44.8 and 40.4%, respectively. The effluent DO concentrations were increased to above 3 and 3.5 mg/L, respectively. Thus, the results in **Con-4** and **Con-5** further proved that intra-membrane air pressure had a significant effect on promoting the removal of NH_4^+-N in the MABR.

The effects of liquid flow velocity on the performance of MABR were investigated by experiments during **Con-4**, **6**, and **7**. The intra-membrane air pressures were kept at 0.08 MPa and other operation conditions were kept the same, the quality of influent were also kept basically the same during **Con-4**, **6**, and **7**.

In **Con-6**, feed flow velocity was increased slightly to 0.2 cm/s. The average removal efficiency of COD in **Con-6** was slightly higher than those in **Con-4**, up to 49.6%. The average removal efficiency of NH_4^+-N was substantially increased, up to 95.5%. After 24 d, effluent COD remained at 150 mg/L and effluent NH_4^+-N was below 5 mg/L. Under visual observations, the biofilm did not appear to shed from hollow fiber membranes, which can explain the biofilm tended to be dense and cohesive under the high flow stream [22,23].

Comparing the experimental results in **Con-4** and **Con-6**, it can be concluded that COD and NH_4^+-N removal could be significantly enhanced by increasing feed flow velocity in the MABR. In **Con-7** (141–160 d), feed flow velocity was increased to 0.05 m/s to further investigate the effect of flow velocity on COD, oil, and NH_4^+-N removal, and the activities of different bacteria in the biofilm. With the increase in feed flow velocity, at the beginning of **Con-7**, effluent COD and NH_4^+-N decreased slightly further, were below 138 and 3 mg/L, respectively. Meanwhile, the average removal efficiency of COD further increased slightly, reached to 57.3 and 95.6%. And this advantage becomes weak during the later of **Con-7**. The removal efficiency of COD was 50.5%. This phenomenon can be explained by that biofilms changed in the later period of **Con-7** under conditions of high velocity result in changing structures that present a significant mass transfer resistance to substrates [21]. However, the MABR performance during **Con-7** was better than that in **Con-6**. Oil removal basically did not change much. These results indicated that MABR tended to preferentially remove NH_4^+-N at high feed flow velocity [24].

3.1. Effects of high SS on MABR performance

A tracking test was conducted on the thirty-sixth day during **Con-1**, with 405 mg/L COD, 29.67 mg/L oil, 76 mg/L SS, and 30.8 mg/L NH_4^+-N at the flow velocity of 0.01 m/s.

As shown in Fig. 3(a), the decline curves of COD, oil, and SS removal rate are steep during the initial 2 h, and then become smoother in the rest of time. The effluent COD, oil, and SS maintained 280, 9.61, and 20 mg/L, respectively. The removal efficiencies of COD, oil, and SS were 30.9, 76.6, and 73.7%, respectively. Fig. 3(b) shows the decline curve of NH_4^+-N removal rate is smooth. The effluent NH_4^+-N was 25.1 mg/L and NH_4^+-N removal efficiency was just 18.5%. Although a high intra-membrane air pressure was provided, the concentration of DO in liquid was substantially low during the experiment.

As shown in Fig. 3(a), during the initial 2 h, the rapid decrease in COD, oil, and SS concentration were almost due to their adsorption on BL. Then, the COD, oil, and SS concentrations decline relatively slow due to the difficulty of adsorption of the residual by BL or the difficulty of diffusion of residual to the inner side of the biofilm to be degraded by micro-organism. As shown in Fig. 3(b), the removal efficiency was low due to insufficient ammonium oxidation. The BL weakened NH_4^+-N transfer from bulk to the inside of biofilm, and thus led to the limitation of the activity of nitrifying bacteria under low NH_4^+-N concentration.

SS consist of non-soluble suspended solids (NSS) and soluble suspended solids (SSS). The volume of NSS is relatively large, which are conducive for oil particles to being adsorbed on the surface of NSS during wastewater circulation in the MABR. NSS easily attach on the outside surface of the biofilm. As more and more such NSS attached, the biofilm was wrapped by the BL. The thicker the BL was, the greater the impact effect on MABR's performance the BL had.

In summary, high SS in influent has negative effects on the performance of MABR. Therefore, it is particularly important to reduce the concentration of SS in influent before wastewater was added into MABR.

3.2. Effects of intra-membrane air pressure on MABR performance

Three tracking tests were conducted to explore the effects of intra-membrane air pressure on COD, oil, and nitrogen removal processes at the seventy-fourth, ninety-third, and ninety-fourth day in **Con-3**–**5**, respectively.

As shown in Fig. 4(a), COD removal rates at 0.12 and 0.08 MPa are higher than that at 0.02 MPa, up to 45.5 and 44.3%, respectively. The decline of COD concentration are relatively fast during the initial 6 h and slow down between the eighth and the twentieth

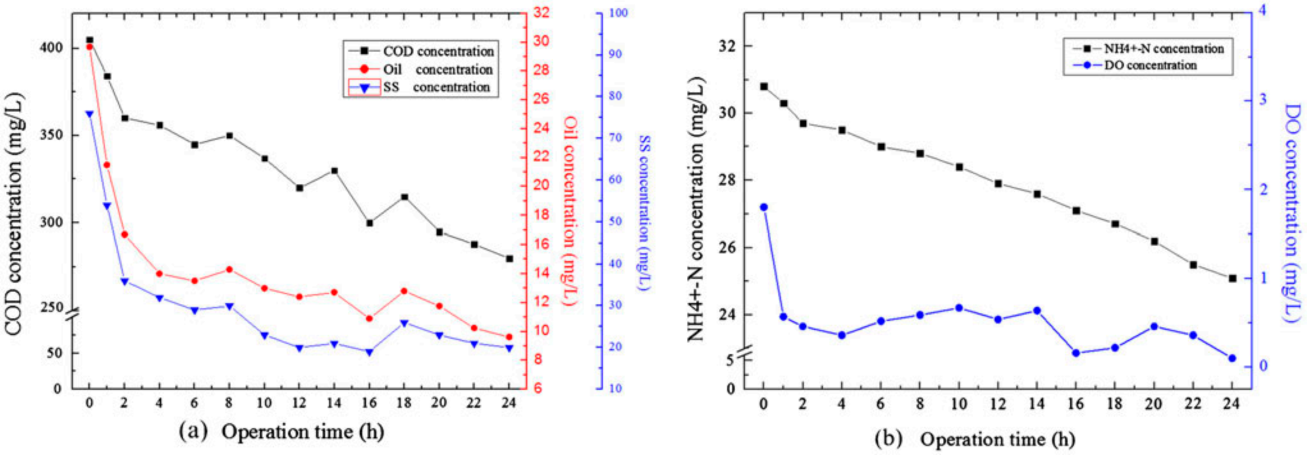

Fig. 3. Variation of COD, oil, DO, and ammonium nitrogen concentration under the condition of the flow velocity of 0.01 m/s and the air intra-membrane of 0.12 MPa. (a) COD, oil, and SS concentrations in the MABR influent and effluent; (b) NH_4^+-N and DO concentrations in the MABR.

hour, and then almost no removal in the rest time. During the initial 8 h, as shown in Fig. 4(f), oxygen permeating into the biofilm through the membrane was almost consumed within the biofilm. The aerobic layer, adjacent to the membranes contains rich oxygen and enough carbon substrate providing a well environment where oxygen is available for the activities of aerobic heterotrophiey; the layer adjacent to the liquid is anoxic, which enhances the activities of denitrifying bacteria and anaerobic heterotrophy. Therefore, the removal rate of COD is enhanced. After 18 h of operation at 0.12 and 0.08 Mpa, the activities of aerobic heterotrophy to reduce COD and nitrifying bacteria to reduce NH_4^+-N were limited due to the limit of available carbon substrate. Anoxic and anaerobic activities were reduced and then stopped in biofilm due to much oxygen permeated the biofilm into liquid making biofilm a aerobic state. And these results the increase in DO concentration in liquid and the decrease in COD removal rate. An endogenous layer for endogenous respiration also existed in biofilm under this condition [18]. The removal efficiency of COD at 0.08 MPa was higher than that at 0.12 MPa, which can be explained by anoxic and anaerobic activities were prematurely limited due to excessive oxygen provided at 0.12 MPa. The limit of oxygen was an important parameter influencing the MABR performance when the intra-membrane air pressure is 0.02 MPa. The activities of aerobic heterotrophy and nitrifying bacteria were limited under low DO concentration and this resulted in low removal efficiencies of COD and NH_4^+-N.

The trend of oil removal rate is basically consistent with the trend of COD removal rate in any of the three aeration pressures. The effluent oil concentrations at 0.1, 0.05, and 0.02 MPa were reduced to about 3 mg/L after 20 h of operation.

In comparison with the removal of organic substrates from wastewater, the nitrification process has relative high oxygen dependence [12]. As shown in Fig. 4(c), NH_4^+-N removal rates at 0.12 and 0.08 MPa are obviously higher than that at 0.02 MPa. At 0.12 MPa, the effluent NH_4^+-N concentration is below 5 mg/L and NH_4^+-N removal efficiency reaches to 91.2%. As shown in Fig. 4(e), nitrate concentration accumulates quickly at 0.12 MPa than those at 0.08 and 0.02 MPa, which demonstrates that high aeration pressure limits the opportunity for denitrification. Fig. 4(d) shows that during the initial 10 h, nitrite concentration accumulates quickly to 5.7 mg/L at 0.02 MPa while it accumulates slowly to 3.5 mg/L at 0.08 MPa. The reason for this phenomenon is that low oxygen concentration facilitates the formation of nitrite, and the activity of nitrite oxidizing bacteria becomes very low when they have to compete for oxygen with ammonia oxidizing bacteria [25]. The accumulate rate of nitrite is the slowest at 0.12 MPa. In any case, nitrite concentration is less than 0.15 mg/L after 16 h of operation.

3.3. *Effects of feed flow velocity on MABR performance*

Three loading rate experiment were conducted to explore the effects of flow velocity on COD, oil, and nitrogen removal processes at ninety-third, one hundred and fortieth, and one hundred and forty-first day in **Con-4**, **6**, and **7**, respectively.

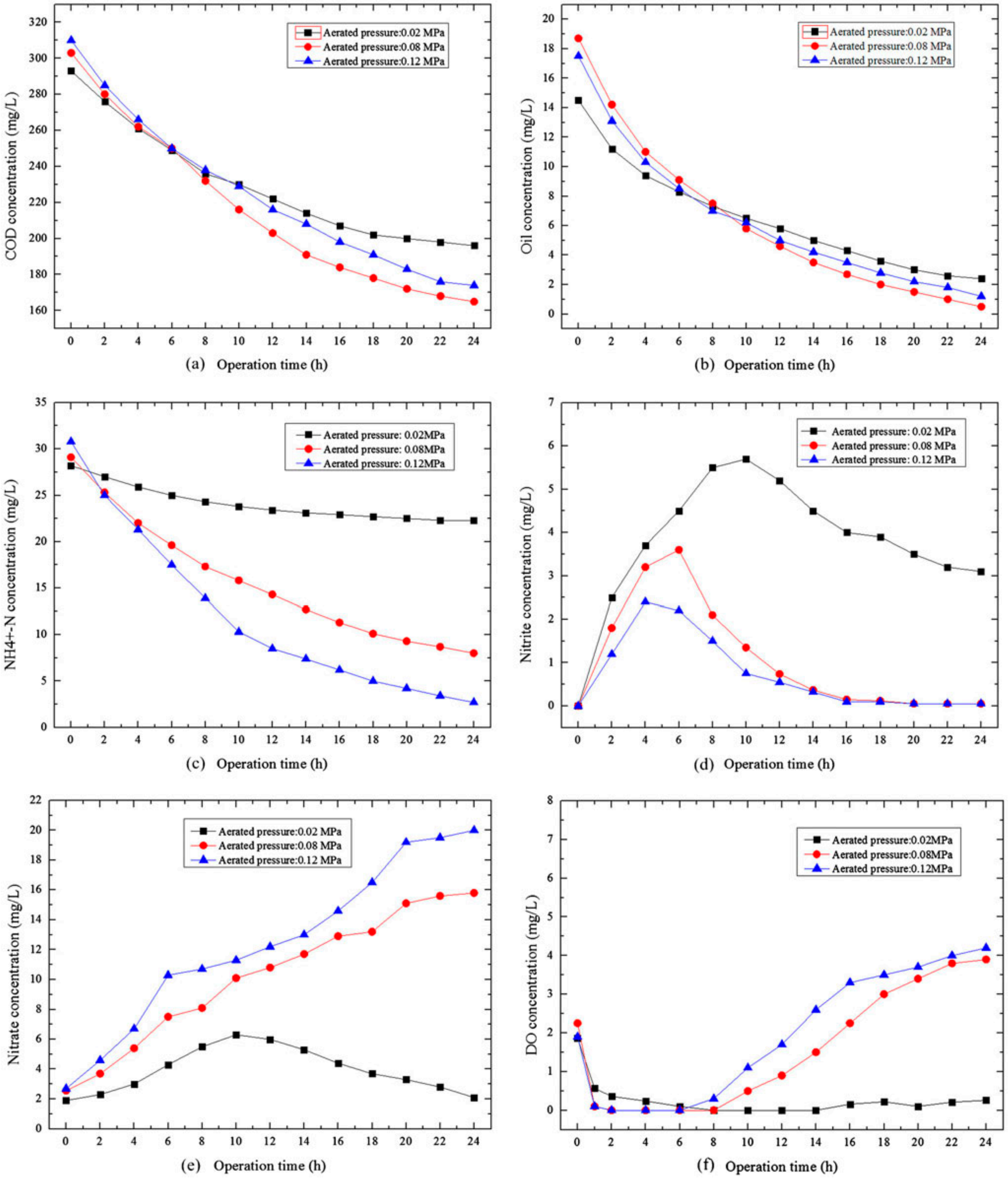

Fig. 4. Variation of COD, oil, NH_4^+-N, nitrite, nitrate, and DO concentrations under different intra-membrane air pressures. (a) COD concentration in the MABR; (b) Oil concentrations in the MABR; (c) NH_4^+-N concentrations in the MABR; (d) nitrite concentrations in the MABR; (e)nitrate concentrations in the MABR; (f) DO concentrations in the MABR.

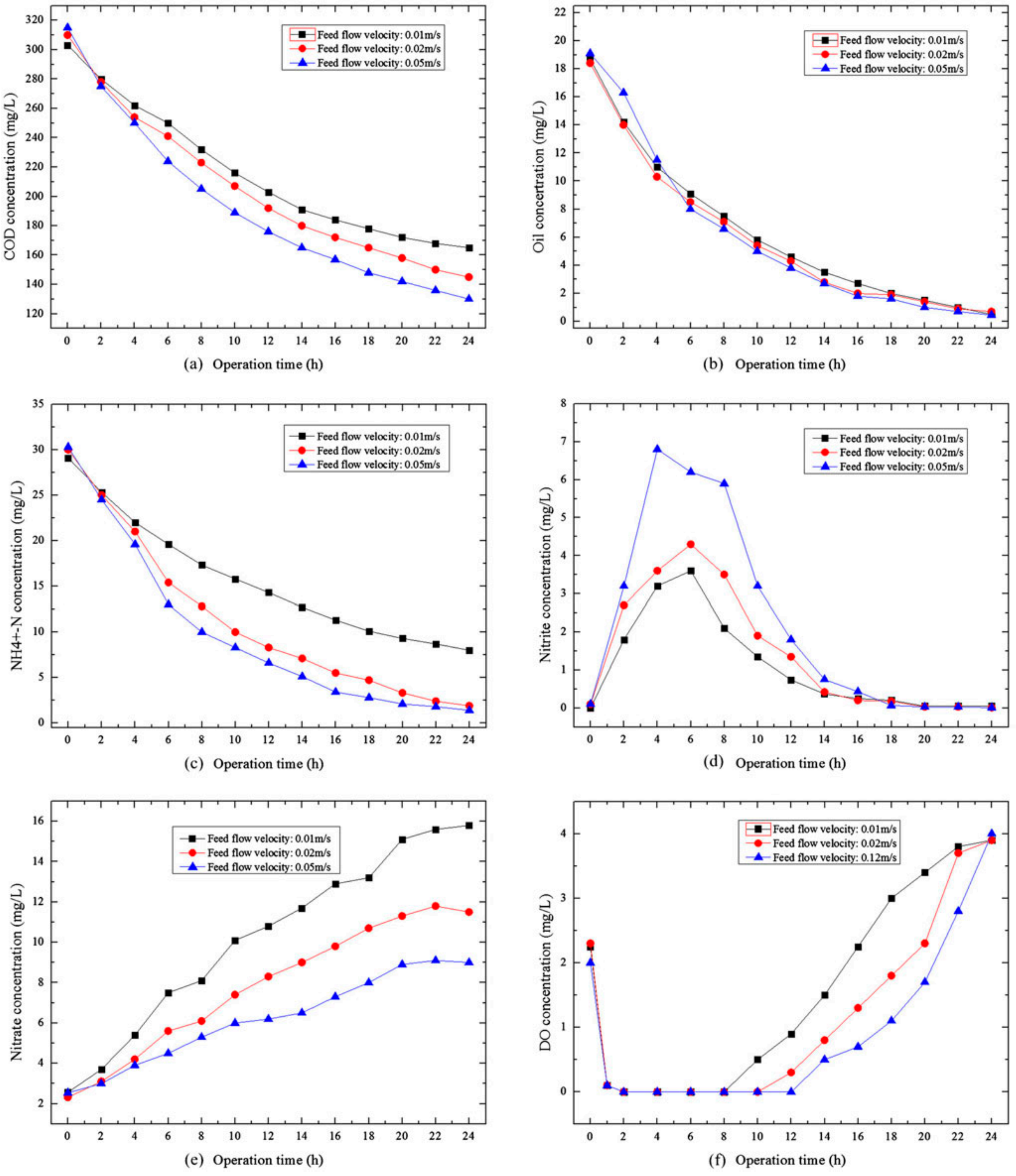

Fig. 5. Variation of COD, oil, NH_4^+-N, nitrite, nitrate, and DO concentrations under different feed flow velocities. (a) COD concentration in the MABR; (b) Oil concentrations in the MABR; (c) NH_4^+-N concentrations in the MABR; (d) nitrite concentrations in the MABR; (e)nitrate concentrations in the MABR; (f) DO concentrations in the MABR.

As shown in Fig. 5(a) and (b), with the increase in feed flow velocity, COD, and oil removal rates were improved in different degrees. At the flow velocity of 0.05 m/s, the removal efficiencies of COD and oil reached 58 and 95%, respectively. The oil concentration decreased to 2 mg/L after operation of 16 h. As shown in Fig. 5(c), NH_4^+-N removal rate at 0.05 m/s flow velocity was significantly higher than that at 0.01 m/s. NH_4^+-N concentration was reduced to 2.1 mg/L and NH_4^+-N removal efficiency reached to 93% at the flow velocity of 0.05 m/s after operation of 20 h. Meanwhile, the NH_4^+-N removal efficiency was just 68% at flow velocity of 0.01 m/s with the identical operation time. The increase in flow velocity enhanced transfer of NH_4^+-N from bulk feed to the inside of biofilm, and thus overcame the limitation of low NH_4^+-N concentration to the activity of nitrifying bacteria.

As shown in Fig. 5(d) and (e), the variation trends of nitrite and nitrate concentrations are different at the different feed flow velocities. During the initial 12 h, nitrite concentration accumulated rapidly, and increased to more than 6.5 mg/L at 0.05 m/s. By contrast, nitrite concentration increased slowly at feed flow velocity of 0.01 m/s with maximum of 3.5 mg/L. The accumulation of nitrate concentration was not obvious under 0.05 m/s after operation of 18 h, the denitrification was terminated (Fig. 5(d)), while nitrification continued and the nitrite formed during nitrification process rapidly and converted to nitrate due to the aerobic state of biofilm and the rich DO concentration in liquid (Fig. 5(f)). This resulted in the decline of NH_4^+-N and the increase of nitrate.

The above results indicate that the MABR process for treating OPWFOOP was also impacted by the feed flow velocity. Higher flow velocity decreases the boundary layer thickness nearby the biofilm [6], and thus increases the concentration gradient in the biofilm [20,26]. The higher concentration gradient of substrates means the greater mass transfer driving force which facilitates organic substrates and NH_4^+-N to transfer into the aerobic and nitrifying zone with high oxygen concentration where nitrification happens in the MABR. Meanwhile, the activities of nitrifying bacteria and aerobic heterotrophy are enhanced. Therefore, the nitrification process is strengthened and the ability of ammonium preferential removal is achieved. When the transfer rate of contaminants is the controlling factor, the increase in mass transfer could enhance the removal rates and efficiencies of NH_4^+-N and COD, which can be seen from the results of Fig. 5 and the comparison of **Con-4**, **Con-6**, and **Con-7** in Fig. 2.

4. Conclusion

A MABR system was developed to investigate the feasibility of OPWFOOP treatment. It is found that simultaneous COD, oil, and NH_4^+-N removal existed in a single reactor. High SS in feed had negative effects on the MABR performance mainly through forming BL on the outside surface of biofilm. The intra-membrane air pressure showed evident effects on the MABR performance mainly through affecting the activities of micro-organism in biofilm. The feed flow velocity also had remarkable effects on the MABR performance mainly through facilitating mass transfer to and in the biofilm. The highest removal rate of COD, oil, and NH_4^+-N achieved as 60.3, 80, and 95%, respectively, with the feed flow velocity of 0.05 m/s and intra-membrane air pressure of 0.08 MPa in this study. This study shows that MABR in treating OPWFOOP has a good prospect. In this study, the structure of biofilm under the influence of flow velocity has not been discussed. Further studies should focus on that and improve COD removal efficiency and system optimization.

Acknowledgments

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