

Positive roles of biofilm during the operation of membrane bioreactor for water reuse

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Received 31 July 2005; accepted 23 December 2005

Abstract

Generally, membrane biofilm have been believed to be minimized during the operation of membrane bioreactor (MBR) for wastewater treatment and reuse. In this study, positive roles of membrane biofilm in the intermittently aerated MBR system were highlighted. During the long-term operation using real domestic wastewater, membrane biofilm played secondary filtration barrier for both low and high molecular weight organic matters. The biofilm on the membrane surface was responsible for the removal of low molecular weight organic matters by the use of easily degradable organic matters (i.e. <1 kDa) as microbial carbon and energy sources during the filtration. Also, track study showed that the significant denitrification took place by the membrane biofilm, proved by the degradation of carbon source and increase of alkalinity such as inorganic carbon. Nevertheless the necessity of further study, this study will give unique insight into positive roles of membrane biofilm, and also continue to aid both fundamental studies and developments of MBR processes.

Keywords: Membrane bioreactor; Biofilm; Denitrification; Secondary membrane; Molecular weight distributions

1. Introduction

In areas of water scarcity, the reuse of wastewater became attractive means as an alternative water source. In many technologies, membrane bioreactor (MBR) has been focused because of its

excellent effluent quality to meet the guidelines of water reuse. Membranes in ultra- or micro-filtration range can achieve the pathogen-free effluent, higher concentration of biomass and longer sludge retention times (SRT), which are beneficial to the low growth bacteria such as nitrifiers [1,2]. Furthermore, high molecular organic compounds, which cannot be degraded by conventional

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Presented at the conference on Wastewater Reclamation and Reuse for Sustainability (WWRS2005), November 8–11, 2005, Jeju, Korea. Organized by the International Water Association (IWA) and the Gwangju Institute of Science and Technology (GIST).

biological systems, do not pass the UF membrane, and are eventually degraded or discharged with the excess sludge [3].

However, the membrane biofouling, which have been regarded as an Achilles heel from the beginning of the membrane technology development, is inevitable in applying MBR although a lot of anti-fouling strategies, such as high cross-flows, chemical dose, vibration, air sparging, membrane modification and the like, have been investigated [4]. Membrane fouling is mainly caused by the attachment of suspended solids and soluble substances on the membrane surface, and influenced by a number of factors relating to the feed water (pH, composition, characteristics, etc.), membranes (geometry, material composition, pore size distribution, etc.) and operation (hydrodynamics, foulant concentration, etc.) [5,6]. Fouling most commonly takes place external to the membrane, forming a dynamic layer at the membrane surface.

At the same time, particle deposition or collection in the cake layers on the membrane surface can play the role of an additional separation bed by capturing or screening materials which would otherwise pass through the membrane pore. This layer may behave as a secondary membrane which removes the smaller foulants before they reach the membrane surface [7]. Davis group studied dead-end microfiltration of various protein mixtures such as lysozyme, ovalbumin and bovine serum albumin (BSA) through yeast cake layers [8–10]. They clearly showed that the yeast formed a cake layer on the membrane surface and acted like a dynamic or secondary membrane, allowing protein monomers to pass through but preventing protein aggregates from fouling the membrane. A membrane-attached biofilm (MAB) process is another example of membrane biofilm. In the MAB systems, the biofilm develops on the outside surface of the membrane and removes nitrate or ammonia by counter diffusion of organic carbon source or dissolved oxygen, respectively [11,12].

In this study, a new aspect of membrane biofouling or roles of membrane biofilm during MBR operation will be focused.

2. Materials and methods

2.1. Reactor and operation

An intermittently aerated MBR was operated using real wastewater over 1 year, alternating anoxic (70 min) and aerobic (110 min) conditions in one cycle (3 h). The permeate flow was ceased during first 40 min of anoxic conditions, and then started at 0.3 m/d of the permeation velocity during the rest of one cycle. The detailed explanation of schematics and operations of the reactor are found in elsewhere [13]. The membrane used in this study was the hydrophilic poly-sulfone membrane with the molecular weight cut-off of 200 kDa suggested by the manufacturer. Average SRT and the concentration of biomass were about 60 dmays and 7000 mg/L, respectively. The suction pressure was controlled to keep the flux from 0.25 to 0.3 m/d and the membrane was chemically cleaned using 5% H₂O₂ solution when the suction pressure became higher than 300 mmHg.

2.2. Samples and measurement

The performance of the reactor was monitored two to three times a week by analyzing the quality of influent, effluent and supernatant of MBR. Additionally, track studies were conducted during two cycles at day 137 and 180 with sampling in every 20 min. Samples were monitored via online (pH and DO) or directly (nitrate and DOC) to minimize the sampling error.

2.3. Analysis of molecular weight distribution and biofilm

The molecular weight distribution (MWD) was evaluated by UF membranes (Amicon, YM series)

with molecular cut-off of 1, 10, 30 and 100 kDa according to the method suggest by Logan and Jiang [14]. As their model, the initial concentration of solute (C_{ini}) could be calculated using Eq. (1):

$$\log C_p = \log(p \times C_{ini}) + (p \times 1) \log F \quad (1)$$

where p = permeation coefficient, C_p = measured instantaneous permeate concentration, C_{ini} = initial bulk concentration of solute, F = fractional reduction in retentate volume.

Both p and C_{ini} can be determined from a linear plot of $\log C_p$ versus $\log F$ by taking the intercept and slope as the $\log(pC_{ini})$ and $(p-1)$, respectively.

3. Results and discussion

3.1. Performance of the MBR

Table 1 summarized the characteristics of influent, effluent and the supernatant of the reactor. While the total organic carbon (TOC) of the influent varied from 156 to 72 mg/L, the effluent TOC always maintained a level below 5 mg/L during the normal operation period, with a lowest value of 1.9 mg/L throughout the study. Meanwhile, the removal efficiencies of nitrogen and phosphorous were reached to 82.2 and 44.1%, respectively, despite the severe fluctuation of influent characteristics. The trans-membrane pressure was gradually increased during the operation and it resulted in chemical cleaning

every 2–3 months. Although the chemical cleaning was effective to remove biofilms on the membrane surface, 100% of the initial flux recovery was not observed, which implied that the irreversible fouling occurred.

3.2. Molecular weight distribution of membrane effluent

As mentioned in previous researches [3], high molecular weight compounds in MBR could be retained and accumulated in the supernatant. In order to investigate the fate of organic matters, the MWD of the MBR supernatant, MBR effluent and the filtrate of supernatant by fresh membrane was examined (Fig. 1).

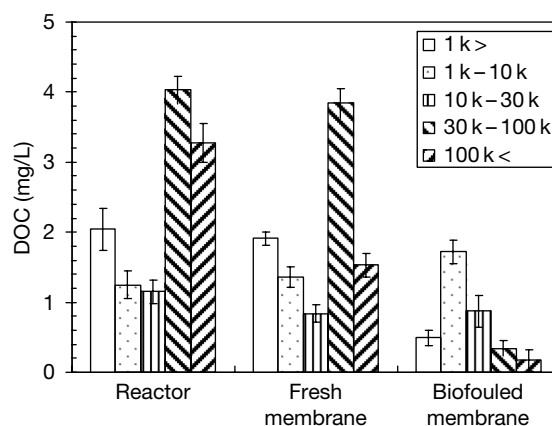


Fig. 1. Molecular weight distributions of filtrates of supernatant by GF/C (reactor), fresh membrane, and biofouled membrane ($n = 2$).

Table 1

Characteristics of influent, effluent and supernatant of the membrane bioreactor (MBR) system

Type		pH	TOC (mg/L)	TN (mg/L)	NH ₄ ⁺ -N (mg/L)	TP (mg/L)
Influent	Max.	7.4	156	77	51	5.0
	Min.	6.7	72	37	19	1.9
	Average	7.1	102	58	41	3.4
Effluent		7.2 ± 0.2	3.6 ± 0.8	10.3 ± 2.9	1.5 ± 0.9	1.9 ± 0.6
Removal efficiency		–	96.5%	82.2%	96.3%	44.1%

The MWD profile of the GF/C and fresh membrane filtrate shows that the membrane can retain high molecular weight organic matters greater than 100 kDa, which is consistent that the molecular weight cut-off of the membrane was 200 kDa. Meanwhile, MWD of MBR filtrate (or filtrate of biofouled membrane) showed that both low (<1 kDa) and high (>30 kDa) molecular weight compounds were effectively retained in the reactor. It is clear that the removal of higher molecular weight organic matters by the biofouled membrane was due to the membrane separation either by membrane pores or by biofilm layer. But the molecular weight cut-off of the membrane used in this study was 200 kDa, which could not separate low molecular matters as low as 1 kDa. As explained in our previous research [3], biofilm on the membrane surface was responsible for the removal of low molecular weight organic matters. Microorganisms in biofilm might use easily degradable organic matters (i.e. <1 kDa) as their carbon and energy source during the filtration. Extra molecular barrier for both low and high molecular weight compounds would be the first positive role of membrane biofilm in MBR process as discussed in previous researches.

3.3. Denitrification by the membrane biofilm

During the experiments, we found that the concentration of nitrate was significantly different between supernatant and effluent of MBR. To examine the mechanism of nitrate reduction during the MBR filtration, we conducted track study during two cycles. In the track study, the concentration of nitrate in the effluent was always lower than that in the reactor (Fig. 2), which implied that denitrification could have taken place during the filtration. Potential removal mechanisms would be (1) Rejection by membrane or biofilm, (2) Aerobic denitrification by attached biofilm, (3) Denitrification during filtration by counter diffusion of nitrate. The

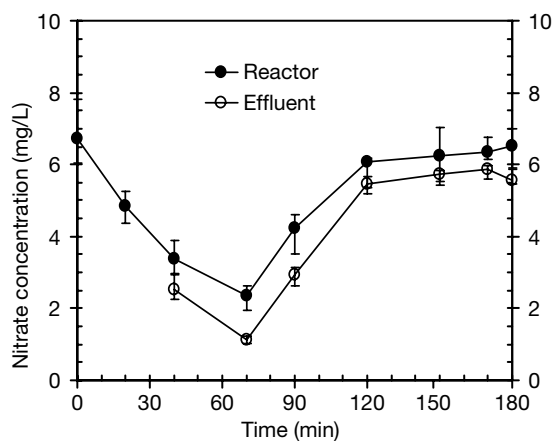


Fig. 2. The nitrate concentration of the supernatant of the reactor and membrane effluent during one cycle ($n = 3$).

first would not be possible because other ions such as Na^+ , Cl^- and NH_4^+ were not varied during the filtration (data not shown). Also, aerobic denitrifiers such as *Thiospaera pantotropha*, *Pseudomonas* sp., *Pseudomonas aureofaciens* and *Alcaligenes faecalis* [15,16] were not found in PCR-DGGE analysis with the same band location for mixed liquor and attached biofilm (Fig. 3). Consequently, denitrification by the MAB would be the most possible mechanism as discussed in related researches of MAB process.

It could be proven in two ways by the combination with Eq. (2), which showed the chemical reaction formula of denitrification process when the wastewater was used as organic sources [17].

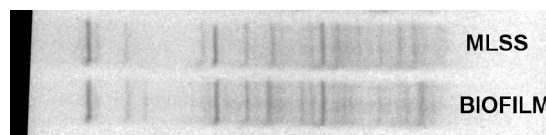
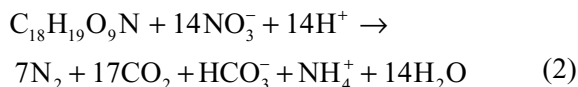


Fig. 3. PCR-DGGE profiles of mixed liquor and attached biofilm in MBR process.

Firstly, the MWDs of effluents from the fresh membrane (i.e. without membrane biofilm) and from the biofouled membrane showed that organic compounds less than 1 kDa were removed during the filtration as discussed above. For denitrification, low molecular or readily biodegradable organic matters [as $C_{18}H_{19}O_9N$ in Eq. (2)] are critical and organic matters less than 1 kDa of the molecular weight could be used as organic sources to denitrify nitrate on or in the membrane biofilm.

Secondly, alkalinity such as CO_2 (aq) and HCO_3^- should be increased during the denitrification as explained in Eq. (2). The variations of inorganic carbon concentration during the track study are shown in Fig. 4. It is worth to note that the concentration of inorganic carbon, or the sum of aqueous CO_2 and HCO_3^- , was increased during the filtration. It is the evidence that anoxic denitrification was taking place.

By numerically integrating the permeation flow rate and concentration data during track studies, it is possible to calculate the mass balance of each constituent at the single cycle. From these calculations, about 53.2 g was entered in the reactor during the cycle: Of them,

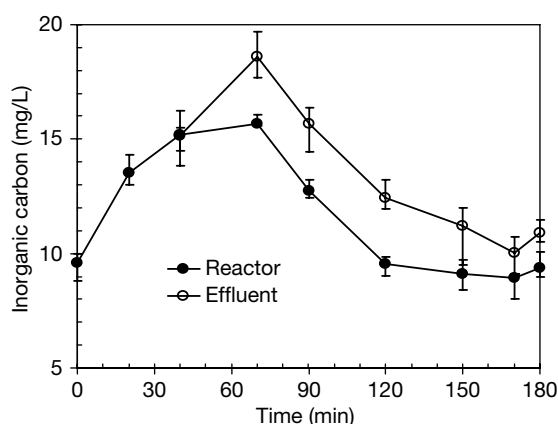


Fig. 4. The inorganic carbon concentration of the supernatant of the reactor and membrane effluent during one cycle.

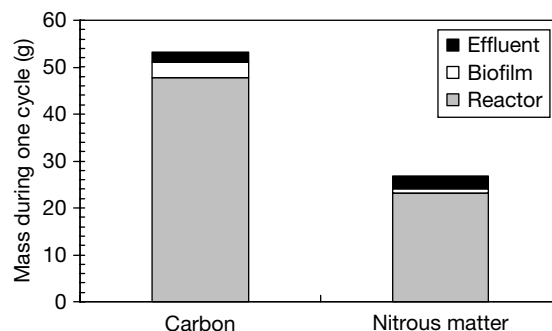


Fig. 5. Mass balance of removed organic carbon and nitrous compounds during one cycle.

47.8 g (89.8%) and 3.3 g (6.3 %) were removed by the mixed liquor and membrane biofilm, respectively, and only 2.1 g (3.9%) of organic carbon were discharged with the effluent. For nitrous compounds, 26.7 g of nitrous compounds (mainly organic nitrogen and ammonia) were introduced into the reactor: 23.1 and 0.93 g were removed by mixed liquor and membrane biofilm, respectively (Fig. 5).

Above results confirmed that MAB did not play only biofouling of membrane, but also positive roles such as molecular barrier for both low and high molecular range, and denitrification of nitrate during the filtration.

4. Conclusions

This research confirms that the membrane biofilm can play negative roles such as the increase of a trans-membrane pressure and the decrease of an effluent flux, as well as positive roles such as secondary barrier for both low and high molecular weight compounds, and attached denitrifying biofilm. Although the exact mechanistic explanation was not completed yet, this study will give unique insight into positive roles of membrane biofilm, and also continue to aid both fundamental studies and developments of MBR processes.

Acknowledgements

This research was supported by the G-7 Project from Ministry of Environment.

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