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Selecting aeration in a PVDF flat-sheet membrane bioreactor for municipal wastewater treatment

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ABSTRACT

Membrane bioreactors (MBRs), which have been widely used in wastewater treatment and reuse, are restricted by membrane fouling and high energy consumption. This study examines the influences of aeration mode and flow rate on membrane fouling to present recommendations for selecting aeration in polyvinylidene fluoride flat-sheet MBR. Results show that continuous aeration (CA) at a constant flow rate can effectively alleviate membrane fouling, although relaxation stage aeration significantly affects the removal of the cake layer. In the short term, increasing the flow rate of CA effectively controls the deposition of suspended solids and alleviates membrane fouling. However, increasing flow rate exerts high shear stress on microbial flocs, thereby reducing microbial size and causing breakage/deflocculation of sludge flocs in the long term. The results indicate that a high flow rate affects sludge properties and the main fouling contributor changes from suspended solids to colloids. Moreover, the threshold ammonia (NH_4^+-N) concentrations of the supernatant and the effluent reveal that removing ammonia can optimize the flow rate of CA.

Keywords: MBR; Aeration; Fouling

1. Introduction

The scarcity of water resources worldwide has created the need to generate new water resources through water reuse. Given the inherent disadvantages of conventional biological treatment processes in effluent quality, footprint, and reliability, membrane bioreactors (MBRs) have gained increasing acceptance and have demonstrated potential in fundamentally advancing the technology and practice of wastewater treatment and reuse [1–3].

However, the current overall cost of MBR treatment is higher than those of conventional biological treatment processes, unless flux is improved and operating costs are diminished [4–8]. That is, developing cost-effective strategies to minimize fouling and finding the best compromise between productivity and costs are necessary. The nature and degree of membrane fouling is influenced by membrane properties, operating conditions, and mixed liquor characteristics. However, pilot-scale experiments and full-scale plants have found that membrane fouling can be effectively controlled by improving membrane properties or operating conditions; successful results can only be

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achieved in laboratories because of the high complexity of mixed liquor characteristics.

Among hydrophobic polymers, polyvinylidene fluoride (PVDF) is the only one that can be used to prepare asymmetric membranes [9–11]. PVDF is a semi-crystalline polymer with a crystalline phase and an amorphous/rubbery phase. The crystalline phase provides thermal stability, whereas the amorphous phase offers flexibility to membranes [12]. In addition, PVDF exhibits outstanding anti-oxidation activity and strong chemical resistance against aggressive reagents [13]. Hence, PVDF membrane has received increasing attention for application in MBR. With respect to external shape, PVDF used for MBR wastewater treatment and reuse is a flat-sheet or hollow-fiber membrane. Compared with a hollow-fiber membrane, a flat-sheet membrane used in MBR has the following advantages. (1) Spacing between membranes can be controlled and, thus, the effective membrane area will not be significantly reduced by the attached sludge. (2) Membranes are convenient to install, and damaged membranes can be uninstalled individually to enable single replacement. (3) Membranes can be easily cleaned physically without backflush.

Aeration is an effective method to control fouling. However, a high aeration rate which is necessary to effectively control fouling, will inevitably result in high energy consumption and operating costs, particularly in submerged MBRs [14]. Thus, operators have to pay attention to increasing aeration rate in view of a costs-saving strategy [15]. Despite significant reductions in energy demand are attainable through operating at lower mixed liquid suspended solids (MLSS) levels and lower fluxes, reducing the flux commensurately increases the required membrane area and thus the capital cost, and decreasing the MLSS increases the sludge production and can also increase the fouling propensity [16]. Therefore, investigating aeration is still significant in alleviating membrane fouling and saving energy. In this study, the influences of aeration mode and flow rate on membrane fouling of PVDF flat-sheet MBRs are investigated to select aeration. Based on the experimental results, recommendations for selecting aeration are presented, which may provide references for designing and operating PVDF flat-sheet MBRs.

2. Materials and methods

2.1. The experimental setup

A pilot-scale MBR with a configuration similar to that of an internal-loop airlift reactor was used in this study for easy scaling up (Fig. 1). A membrane

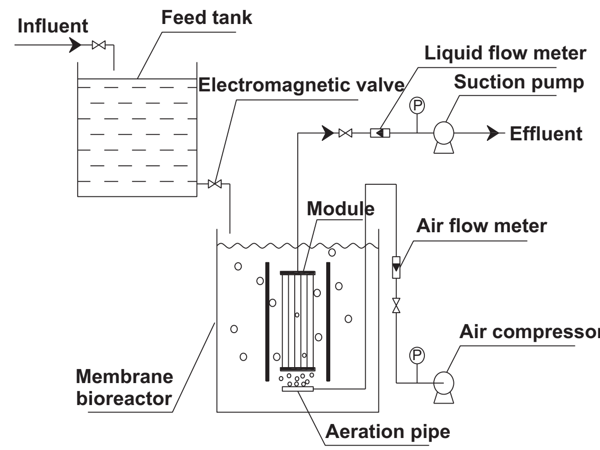

Fig. 1. Schematic of the experimental system setup.

module separated the bioreactor into a riser zone and two down-comer zones that were connected by two bottom flow channels and two top flow channels. The ratio of the total area of the down-comer zones to that of the riser zone was 2:1. The module was built with five PVDF flat-sheet membranes arranged in parallel, with defined spaces between each plate. Table 1 lists the specifications of the PVDF flat-sheet membranes used in this study.

The influent was taken from the feed tank and drawn to the bioreactor by an electromagnetic valve that was controlled by a water-level sensor to maintain a constant water level in the bioreactor over the experimental period. Aeration, which was achieved with an air compressor and introduced through a perforated pipe with a pore size of 3 mm beneath the membrane module, can induce mixing in the bioreactor, remove particles deposited on the membrane surface, and provide oxygen for biomass growth. Given that intermittent operation can improve membrane fouling, this study was performed in an intermittent mode. Effluent was intermittently drawn from the outside to the inside of the membrane by a peristaltic pump that was automatically controlled by a programmable logic controller (OMRON Co., Ltd, Shanghai, China). Filtration and relaxation times were 8 and 2 min, respectively.

2.2. Wastewater characteristics

To reduce the effects of the influent to the MBR, synthetic "municipal" wastewater was used throughout the experiment period. The composition and concentration of wastewater were listed in Table 2. Sodium bicarbonate ($NaHCO_3$) was used as buffer to maintain a constant pH and support nitrification.

Table 1
Specifications of the PVDF flat-sheet membrane used in the present study

Manufacturer	Shanghai SINAP Membrane Science & Technology Co., Ltd., China
Model number	SINAP-10
Configuration	Flat sheet
Material	PVDF
Width × height × thickness	220 × 320 × 6 mm
Weight	0.4 kg
Effective surface area	0.10 m^2
Nominal pore size	0.10 μm
pH resistance range	3–12
Capacity	16.7–25 L/(h m^2)

Table 2
Component and concentration of synthetic "municipal" wastewater (mg/L)

Component	Concentration	Component	Concentration
Glucose	267.0	Peptone	17.0
Soluble starch	267.0	Potassium dihydrogen phosphate	27.0
Sodium bicarbonate	233.0	Calcium chloride	3.0
Ammonium chloride	83.0	Magnesium chloride	3.0
Ferric chloride	3.0	Manganese sulfate	3.0

Before being fed to the MBR, the influent was stabilized at room temperature for 1 d. The parameters of the synthetic "municipal" wastewater are listed in Table 3.

2.3. Operating conditions

The experiment was divided into four stages.

The first stage involved starting up and stabilizing the MBR. The MBR was initially inoculated with returned activated sludge from a municipal wastewater treatment plant. The sludge was acclimatized to the substrate until the MBR reached a steady state. Approximately 90% chemical oxygen demand (COD) removal was achieved, with COD concentration in the effluent being lower than 50 mg/L. Solid retention time was maintained at 20 d during the operating period.

Table 3
Parameters of the synthetic "municipal" wastewater

Parameter	Unit	Average	Range
COD	mg/L	447.0	404.5–489.7
NH_4^+-N	mg/L	13.70	12.58–14.91
Total phosphorus	mg/L	3.92	3.46–6.17
Turbidity	NTU	36.73	26–53
pH	–	7.50	6.70–7.94
Temperature	℃	24.0	21.0–25.5

The second stage of investigation involved selecting the aeration mode. Flow rate, time, and position are three parameters of aeration that affect membrane fouling. Aeration flow rate and time, which are connected, affect energy consumption more significantly than aeration position. According to the intermittent operation period of the peristaltic pump, aeration time can be divided into continuous, filtration stage (8 min), and relaxation stage (2 min) aeration. To select aeration mode, transmembrane pressure (TMP) trends —with the following operation periods: relaxation time: 2 min; filtration time: 8 min—were monitored under an aeration flow rate of 0.7 m^3/h and a membrane flux of 28.32 L/(h m^2).

The third stage was performed to assess the properties of the PVDF flat-sheet MBR by changing aeration flow rate. The result obtained in the second stage was applied. Aeration flow rate (0.45, 0.60, 0.75, and 0.90 m^3/h) was adjusted using a rotameter. Membrane flux was 14 L/(h m^2), and hydraulic retention time was controlled at 10 h. Membrane performance was evaluated based on the critical flux (CF), microbial size, and filtration resistance of different mixed liquor fractions. The biological treatment of wastewater and effluent quality were evaluated in terms of COD and ammonia nitrogen (NH_4^+-N).

Finally, recommendations were provided according to the results.

2.4. Analytical methods

TMP was monitored using a pressure gauge set between the PVDF flat-sheet membrane module and the peristaltic pump. CF was measured via the "TMP-step" method [17]. Flux was calculated as effluent flow rate divided by membrane filtration area. Effluent flow rate was monitored via a volumetric method. All PVDF flat-sheet membranes with an effective filtration area of 0.10 m^2, which were used to determine CF, were located in the riser. Separation and resistance calculation of different mixed liquor fractions (solutes, colloids, and suspended solids) were performed according to Bae and Tak [18]. Microbial size and species were observed through an electron microscope (Scope A1, Zeiss, Germany). Particle size distribution of colloids in the supernatant was analyzed through a particle size analyzer (NanoZS90, Malvern Instruments Ltd., British). COD and NH_4^+-N were determined via a spectrometric method at wavelengths of 610 and 440 nm with a 5B-3B instrument (Lanzhou Lianhua Environmental Technical Co., Ltd., Lanzhou, China). All analyses were performed in triplicate for each sample. The deviation of each measured parameter for each sample was less than 10%.

3. Results and discussion

3.1. Selecting aeration mode

As shown in Fig. 2, TMP increases with time in each aeration mode, thus indicating that membrane fouling gradually deteriorates. However, the level of TMP increase exhibits significant differences under different aeration modes. The TMP value of continuous aeration (CA) is always lower than those of filtration stage aeration (FA) and relaxation stage aeration (RA), with RA exhibiting the highest TMP value. This finding indicates that CA is the most effective mode for alleviating membrane fouling, whereas FA is the worst mode.

Comparing FA with CA, both TMP increase rates are nearly identical, and the only difference is the initial TMP value. This phenomenon can be explained as follows. The cross-flow velocity of the membrane surface is constant in CA mode, whereas it can be divided into three stages (zero, increasing, and stable stages) in FA mode. The zero stage of cross-flow velocity starts in the relaxation phase of the peristaltic pump (2 min), increases, and finally reaches a stable value in the filtration phase of the peristaltic pump

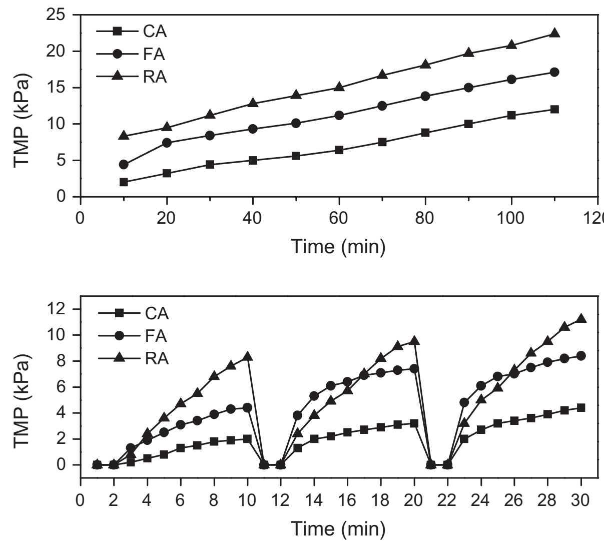

Fig. 2. Changes in TMP with time at different aeration modes (aeration intensity = 0.7 m^3/h, flux = 28.32 L/(h m^2)).

(8 min). The cake layer rapidly develops in the zero and increasing stages. Therefore, the same cross-flow velocity cannot completely remove the accumulated pollutants in the stable stage although it can mitigate further migration of pollutants from mixed liquor fractions to the membrane surface. The result of this process is reflected by a higher TMP at the end of the first operation period (10 min) in FA compared with that in CA. The process is then repeated, and the TMP of the next period always increases based on that of the previous period.

The severity of membrane fouling could increase because aeration time is only 2 min in RA mode. However, TMP increase rate in RA is slightly lower than that in FA at the beginning of filtration phase, thus indicating that RA has a significant effect on removing the cake layer because of the absence of drag force generated by filtration.

In summary, aeration mode significantly affects the rate and extent of membrane fouling along with energy consumption. CA can effectively alleviate membrane fouling at a constant flow rate, although RA exhibits a significant effect on removing the cake layer. Therefore, the following experiment was designed to assess the effect of flow rate on CA mode.

3.2. Determining CF

Based on the "TMP-step" method, CF was studied at flow rates of 0.9, 0.75, 0.60, and 0.45 m^3/h. In determining CF, only a range can be obtained rather than an exact value because the precision of a peristaltic pump is limited and the step increments of flux are not infinitely small. The result is clearly demonstrated in Fig. 3. The interval of CF increased approximately linearly with flow rate. A high aeration flow rate resulted in numerous large bubbles and drastic turbulence of the gas–liquid two-phase flow, which induced highly variable large shear rates against the flat-sheet membrane surface to impede particle deposition. That is, the particles will not be deposited on the membrane surface theoretically when cross-flow velocity is sufficient to offset the opposing permeate velocity.

3.3. Microbial size of activated sludge

The results of the previous two sections are based on short-term experiments and can be summarized as follows: the increase in flow rate can effectively mitigate fouling in CA by fixing sludge characteristics.

In the MBR system, the activated sludge must be circulated at a high flow rate to maintain the suspension and prevent the deposition of foulants on the membrane surface. This case is not conducive to the growth of large protozoa and rotifers for long-term operations; thus, differences in microbial size may exist between the MBR and the conventional activated sludge process (CASP). When flow rate was set to 0.9 m^3/h, several rotifers were observed actively preying on free bacteria and organic particles under the Scope A1 electron microscope (Fig. 4). In addition, the microbial size of multicellular rotifers exhibits no difference between MBR sludge and CASP sludge (Fig. 5), thus indicating that the survival of protozoa (e.g. vorticella) is affected by a strong shear force. Moreover, the microbial size of rotifers can also be affected when flow rate is increased further. Thus, flow rate should be selected by considering the biological activity with a direct effect on biological wastewater treatment.

3.4. Fouling contributions of sludge fractions at different flow rates

The flux-declining behavior of sludge fractions was investigated to reveal the membrane fouling mechanisms of each sludge suspension at different flow rates. Sludge samples at a flow rate of 0.9 m^3/h were fractionated. The three fractions (suspended solids, colloids, and solutes) were successively filtered by 500 mL Filter System (0.22 μm PES, Corning Incorporated, USA) under vacuum pressure. Under constant pressure, the membrane flux was recorded vs. time and the results calculated were presented in Fig. 6 and Table 4. The membrane permeability of each fraction gradually decreased with time, until finally reaching a steady state. Among which, the resistance value of the supernatant was relatively close to the value of the mixed liquor, thus indicating the minimal contribution of suspended solids on membrane fouling. By contrast, the significant difference in resistance values between the supernatant and the solutes indicates that colloids are major contributors to membrane fouling.

Table 5 illustrates that the main fouling contributor changed from suspended solids to colloids as flow rate increased. A high flow rate can effectively control the deposition of suspended solids and improve membrane flux at the same permeation drag, as discussed in Section 3.2. However, the results in the aforementioned section show that this phenomenon can cause

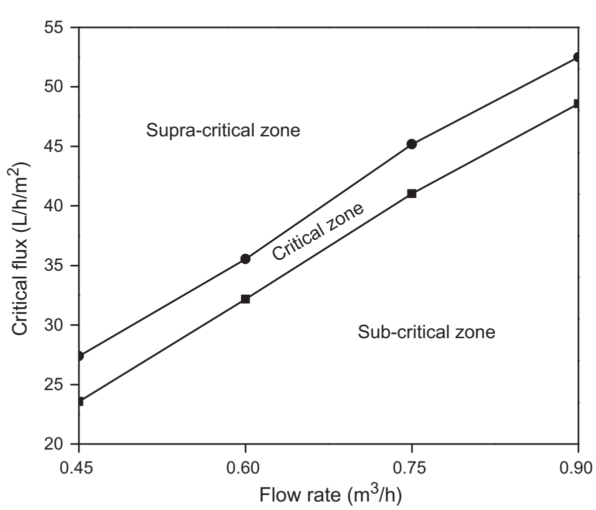

Fig. 3. Variations of CF with aeration flow rate.

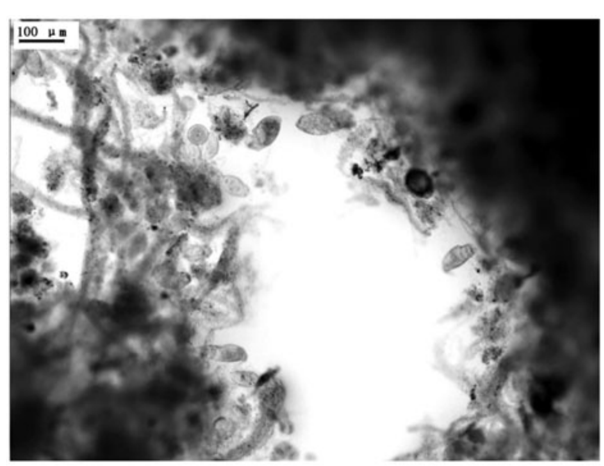

Fig. 4. Microbial species in the MBR.

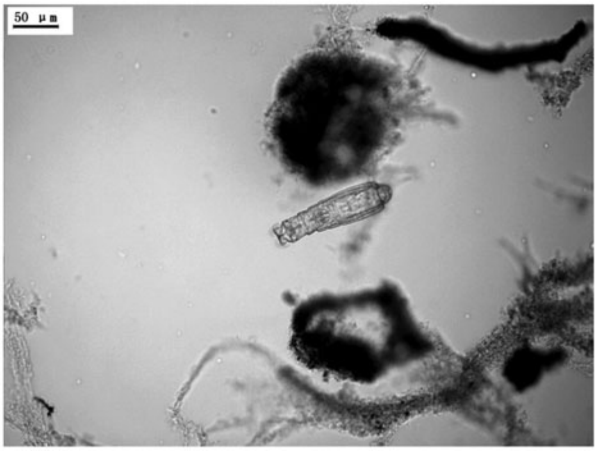

Fig. 5. Sludge floc and rotifer size.

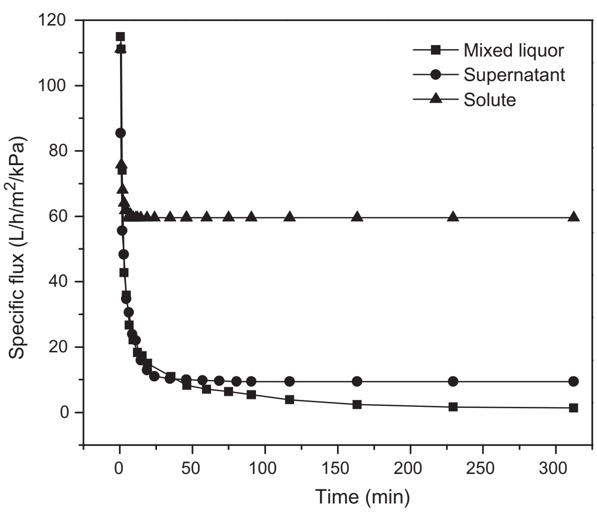

Fig. 6. Variations of specific flux with time during filtration of various sludge fractions.

breakage/deflocculation of sludge flocs during long-term operations. Moreover, colloids that adhere to the outer surface and to the intercellular space of microbial cells are released to the mixture liquor. Therefore, these additional colloids may block membrane pores, form a gel layer on the membrane surface, or embed into the cake layer through permeation drag as flow rate increases. Fig. 7 shows particle size distribution of colloids in the supernatant. It clearly reveals that membrane pores (0.10 μm) can be blocked more easily by colloids than by solute adsorption or suspended solid deposition.

The characteristics of the sludge mixture significantly affect the mechanism and extent of membrane fouling. When the sludge supernatant has a considerable amount of colloids, the contribution of colloids or solutes on membrane fouling becomes the controlling factor. When the relative concentration of these substances in the sludge suspension is small, their contribution to membrane fouling decreases correspondingly [19]. The results of this study indicate that a high flow rate can affect the properties of sludge and the relative contribution of each fraction on membrane fouling. This technique is effective for mitigating macroscopic fouling. However, a high flow rate can also cause serious microscopic fouling of colloids because additional colloids are embedded into membrane pores or deposited onto membrane surfaces by permeation drag. Moreover, these colloids cannot be easily removed by aeration because of their lower back-transport velocity compared with that of suspended solids.

3.5. Removing COD and NH_4^+-N

Soluble, effluent, and colloidal COD concentrations at different flow rates are shown in Fig. 8. The range of effluent COD concentration was from 2.57 to 8.32 mg/L, regardless of the soluble COD concentration in the bioreactor. However, the increase in colloidal COD caused by the breakage/deflocculation of sludge flocs and the release of colloids according to the results in Section 3.2 resulted in high soluble COD concentration.

Nitrogen can be removed by both biomass assimilation and the biological nitrification–denitrification process. However, cell assimilation was estimated to remove approximately 15–20% of influent total nitrogen (TN) concentration, thus demonstrating that the nitrification–denitrification process contributed more

Table 4
Resistance of different mixed liquor fractions at a flow rate of 0.9 m^3/h

Sludge sample	R_t (m^{-1})	R_m (m^{-1})	R_t–R_m (m^{-1})
Mixed liquor	21.15×10^{10}	1.25×10^{10}	$= R_{SS} + R_{col} + R_{sol} = 19.9 \times 10^{10}$
Supernatant	16.55×10^{10}	1.25×10^{10}	$= R_{col} + R_{sol} = 15.3 \times 10^{10}$
Solute	4.03×10^{10}	1.25×10^{10}	$= R_{sol} = 2.78 \times 10^{10}$

Table 5
Filtration resistance and relative fouling contribution of each fraction at different flow rates

Flow rate (m^3/h)	Resistance ($\times 10^{10}$ m^{-1}) and resistance ratio (%)			
	R_{SS}	R_{col}	R_{sol}	R_t
0.9	4.6 (23.12)	12.52 (62.91)	2.78 (13.97)	19.9
0.75	5.53 (31.78)	9.92 (57.01)	1.95 (11.21)	17.4
0.6	9.23 (45.92)	7.76 (38.61)	3.11 (15.47)	20.1
0.45	7.62 (48.23)	4.17 (26.39)	4.01 (25.38)	15.8

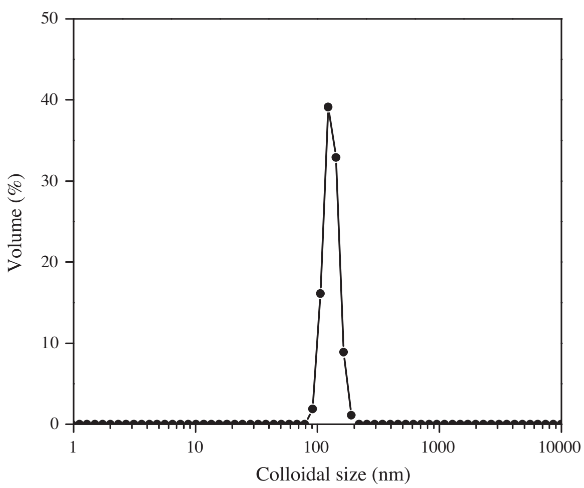

Fig. 7. Particle size distribution of colloids in the supernatant.

significantly to TN removal efficiency than biomass assimilation [20]. Simultaneous nitrification and denitrification (SND) process relies on concurrent aerobic NH_4^+-N oxidation and anoxic denitrification. Fig. 9 shows the NH_4^+-N concentrations of the supernatant and the effluent at different flow rates. In the case of these flow rates, removing NH_4^+-N was accomplished in the bioreactor. This result indicates the occurrence of SND in the MBR because the dissolved oxygen gradient occurred between the riser zone and the down-comer zones of the MBR, thus promoting the activity of nitrifiers and denitrifiers. Meanwhile, the dissolved oxygen gradient also occurred in micro-scale sludge flocs. Thus, the bioreactor structure and sludge flocs create the conditions for nitrification and denitrification.

As shown in Fig. 9, changes in NH_4^+-N concentration in the supernatant and the effluent at different flow rates were similar. NH_4^+-N concentration decreased initially, and then increased with increasing flow rate. When flow rate <0.75 m^3/h, the reduced dissolved oxygen can inhibit the nitrification process.

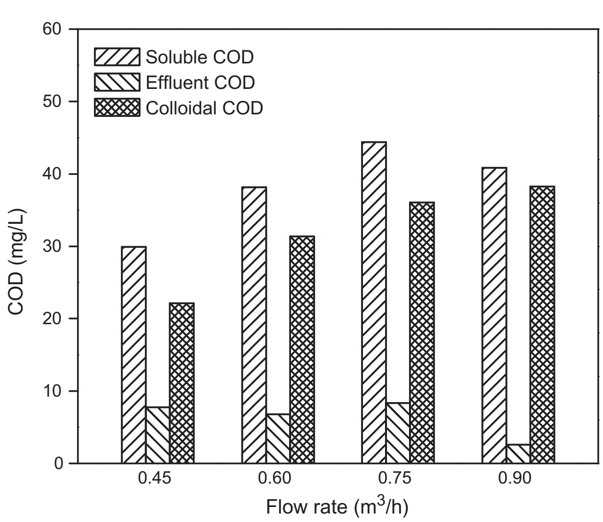

Fig. 8. Concentrations of soluble COD, effluent COD, and colloidal COD at different flow rates.

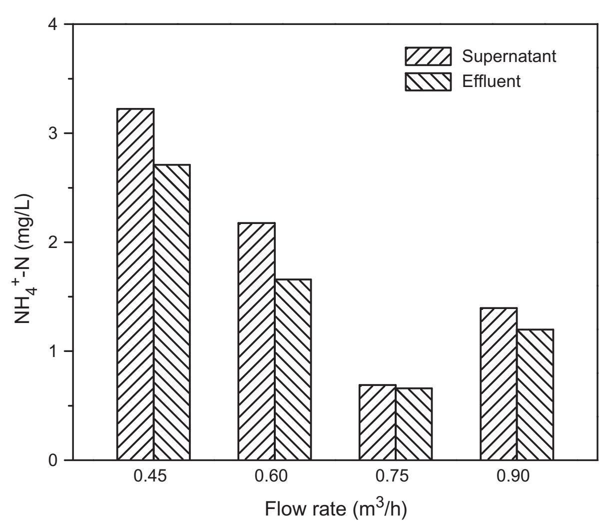

Fig. 9. Supernatant NH_4^+-N and effluent NH_4^+-N at different flow rates.

When flow rate is >0.75 m^3/h, the increased flow rate can theoretically improve the nitrification process, and effluent NH_4^+-N concentration can be maintained at a fairly low level. However, actual NH_4^+-N concentration in the effluent is elevated as flow rate increases because the high shear stress generated by aeration shortens the retention time of nitrifiers in the aerobic riser zone, and thus, microbial biomass in the aerobic/anoxic zone is diluted accordingly. Thus, the activity of nitrifying bacteria is affected at high flow rates. The results suggest a moderate flow rate of 0.75 m^3/h for this case.

3.6. Recommendations for selecting aeration

Based on the experimental results obtained in this study, comprehensive effluent quality, membrane fouling, and energy consumption should be considered in selecting aeration mode. The following recommendations are presented.

(1) Intermittent aeration saves energy but cannot guarantee biological wastewater treatment and cannot alleviate membrane fouling. Thus, an inverter can be used to change flow rate flexibly and enhance aeration in the relaxation stage of the pump according to the results presented in Section 3.1. Moreover, an advanced aeration protocol is currently pulsed aeration.

(2) When CA is applied, micro-organisms in the MBR should exist in the form of attached growth instead of suspended growth to avoid the breakage/deflocculation of sludge flocs and the release of colloids, which can cause serious fouling. Aeration flow rate can then be optimized according to biological wastewater treatment.

4. Conclusions

This study presents recommendations for selecting aeration by investigating the influences of aeration mode and flow rate on membrane fouling of PVDF flat-sheet MBR. The following conclusions can be drawn.

(1) CA can effectively alleviate membrane fouling at a constant flow rate, although RA has a significant effect on removing the cake layer.

(2) In the short term, increasing the flow rate of CA effectively controls the deposition of suspended solids and alleviates membrane fouling. However, increasing flow rate exerts a high shear stress on microbial flocs, thereby reducing microbial size and causing breakage/deflocculation of sludge flocs in the long term. The results indicate that a high flow rate affects the properties of sludge, and the main fouling contributor changes from suspended solids to colloids.

(3) The threshold ammonia (NH_4^+-N) concentrations of the supernatant and the effluent reveal that removing ammonia can optimize the flow rate of CA.

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