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Control of membrane fouling during hyperhaline municipal wastewater treatment using a pilot-scale anoxic/aerobic-membrane bioreactor system

Jingmei Sun[1], Jiangxiu Rong[1], Lifeng Dai[1], Baoshan Liu[1,2], Wenting Zhu[1,*]

1. School of Environmental Science and Engineering, Tianjin University, Tianjin 300072, China. E-mail: jmsun@tju.edu.cn
2. CNOOC Tianjin Chemical Research & Design Institute, Tianjin 300131, China

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Abstract

Membrane fouling limits the effects of long-term stable operation of membrane bioreactor (MBR). Control of membrane fouling can extend the membrane life and reduce water treatment cost effectively. A pilot scale anoxic/aerobic-membrane bioreactor (A/O-MBR, 40 L/hr) was used to treat the hyperhaline municipal sewage from a processing zone of Tianjin, China. Impact factors including mixed liquid sludge suspension (MLSS), sludge viscosity (μ), microorganisms, extracellular polymeric substances (EPS), aeration intensity and suction/suspended time on membrane fouling and pollution control were studied. The relationships among various factors associated with membrane fouling were analyzed. Results showed that there was a positive correlation among MLSS, sludge viscosity and trans-membrane pressure (TMP). Considering water treatment efficiency and stable operation of the membrane module, MLSS of 5 g/L was suggested for the process. There was a same trend among EPS, sludge viscosity and TMP. Numbers and species of microorganisms affected membrane fouling. Either too high or too low aeration intensity was not conducive to membrane fouling control. Aeration intensity of 1.0 m^3/hr (gas/water ratio of 25:1) is suggested for the process. A long suction time caused a rapid increase in membrane resistance. However, long suspended time cannot prevent the increase of membrane resistance effectively even though a suspended time was necessary for scale off particles from the membrane surface. The suction/suspended time of 12 min/3 min was selected for the process. The interaction of various environmental factors and operation conditions must be considered synthetically.

Key words: hyperhaline municipal sewage; anoxic/oxic membrane bioreactor; membrane fouling control; relationship of various factors

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Introduction

Membrane bioreactor (MBR) is the combination of membrane separation technology and biological treatment. It has many advantages, such as high mixed liquor suspended solids, high efficiency, low sludge production and small area requirement. These advantages make the more extensive application of MBR in wastewater treatment. However, membrane fouling is a major bottleneck of the MBR technology. It causes the flux decline and the increase frequency of membrane replacement and cleaning. Thus, the operation costs of MBR system increased significantly. Therefore, the problems of membrane fouling must be solved to expand the large-scale practical application of MBR.

Membrane fouling is caused by the physical and chemical effects among the particles, colloidal particulates or solute molecules in processed materials, by that some solute concentration exceeds its solubility because of concentration polarization on membrane surface, and by that membrane pore sizes become smaller and blocked due to adsorption and deposition of materials on the membrane surface or in the membrane holes by the mechanical action. In general, the fouling occurring in membrane bioreactors is attributable to three aspects: sludge particle deposition, adhesion of macromolecules to the membrane surface, and pore clogging by small molecules. Thus, membrane total resistance (R_t) consists of the intrinsic membrane resistance (R_m), the resistance of membrane pore blocking (R_p) and the resistance of membrane surface cake layer (R_c). R_c was reported as a main contributor to R_t in MBR processes (Lee et al., 2001; Le-Clech et al., 2006). Formation of cake layer was mainly caused by membrane interception. Concentration polarization existing on the membrane surface results in the accumulation of organic molecules and colloid molecules near the membrane surface. When the accumulation reaches to the threshold, a gel layer will form and result in particle deposition and cake layer formation. Contribution of suspended solids, colloidal materials and dissolved substances to membrane

* Corresponding author. E-mail: tjuzwt@sina.com

filtration resistance was 65%, 30% and 5%, respectively (Defrance et al., 2000).

Membrane fouling led to the decrease of flux and the change of separation characteristics (Wan and Wu, 2003). Many factors affect the membrane fouling. The measures to address the membrane fouling are mainly improvement of the sludge mixture properties (Chang and Kim, 2005), pretreatment of water (Wang et al., 2010) and control of operation conditions in membrane separation (Shin and Kang, 2003). Factors affecting sludge accumulation on membrane surfaces were investigated based on the Orthogonal Designed Experiment, e.g., mixed liquid sludge suspension (MLSS), aeration intensity, membrane flux and suction time and non-suction time (Gui et al., 2002).

Many studies documented the impact of single factor or a couple of factors on the membrane fouling. Analyses of the linkages between various factors were seldom reported. In this research, a pilot scale anoxic/oxic-membrane bioreactor (A/O-MBR, 40 L/hr) was used to treat the hyperhaline municipal sewage from a processing area. Optimum operating conditions were investigated to reduce the membrane fouling and to ensure the optimization of operation effects. Impact factors including MLSS, sludge viscosity, microorganisms, aeration intensity and suction/suspended time on membrane fouling and pollution control were investigated, and the correlations among various factors of membrane fouling were also analyzed.

1 Materials and methods

1.1 Raw wastewater quality

Raw wastewater was collected from municipal sewer of a processing zone in Tianjin, China. It consists of approximately 80% industrial wastewater and 20% domestic wastewater. Raw wastewater quality fluctuated in a relatively large range during investigation period, as shown in Table 1.

1.2 Experimental equipment and process

Experimental equipments located in a recycled wastewater treatment plant of a processing zone of Tianjin, China (briefly K plant). The experimental equipments and technical process are shown in Fig. 1. The inoculated sludge of the bioreactor came from the returned sludge of aerobic zone of the third MBR in K plant. After more than one-month cultivation, domestication and stabilization of the A/O MBR system, the experimental investigation started from 24 August, 2009 to 24 December, 2009, totally 123 days.

The treatment capacity of the A/O MBR system was 40 L/hr. Raw wastewater of the system was pumped to a water store tank after going through the bar screen and grit chamber of the K plant, and was further pumped to an elevated tank with heater to keep the influent water above 10°C before flowing into A/O MBR system. The influent water of anoxic zone (A zone, being equipped with stirrer) of A/O MBR was mixed with back flow nitrification fluid from aerobic zone (O zone, being equipped with

Table 1 Raw wastewater quality

Index	Range	Average
COD_{Cr} (mg/L)	42.5–615.1	189.4
Total organic carbon (TOC) (mg/L)	19.93–101.20	40.69
Total N (mg/L)	11.1–41.4	27.6
Ammonia nitrogen (mg/L)	1.7–36.4	16.4
Total P (mg/L)	1.6–7.4	4.7
Salinity (mg/L)	882–1310	1024.6
pH	6.88–9.02	7.83
Suspended solids (SS) (mg/L)	34–294	164

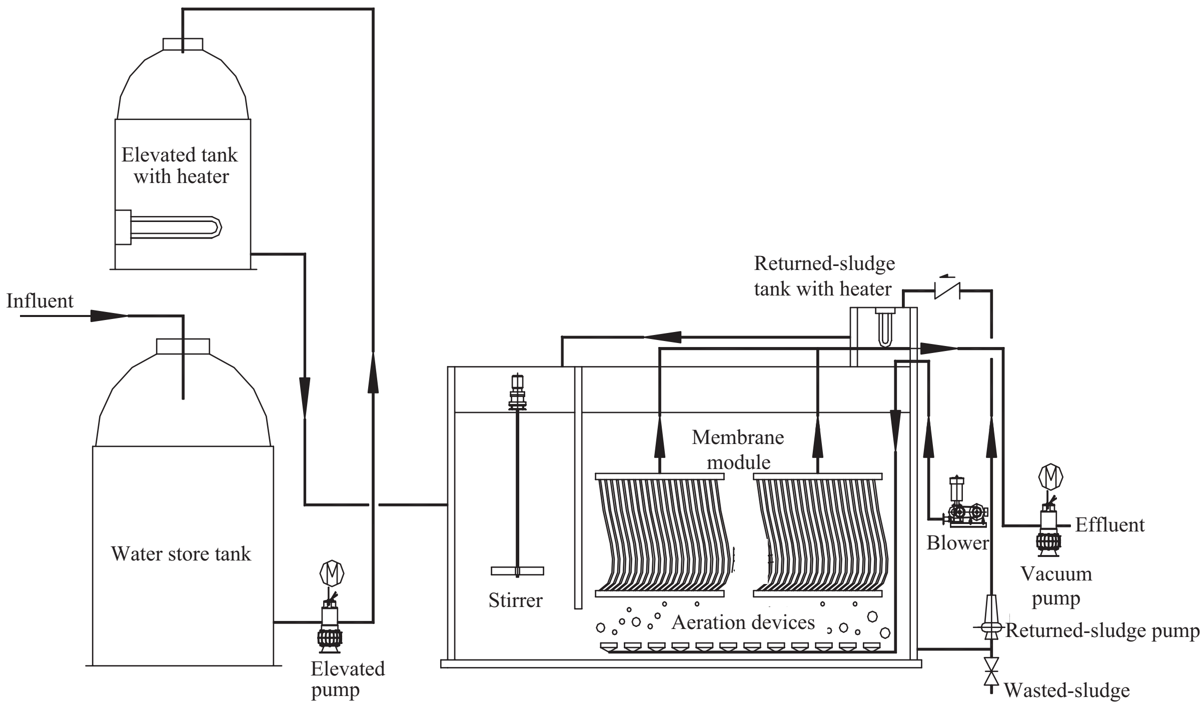

Fig. 1 Experimental equipments and technical process.

membrane module and aeration devices) to denitrify before flowing to O zone. The effluent was drawn out through the membrane modules intermittently by a vacuum pump. Discharge sludge pipe in the bottom of the A zone was designed to connect with returned sludge pump and wasted sludge valve. When the temperature of returned sludge was below 10°C, the heater in the returned-sludge tank was started up.

Experimental devices of the A/O MBR system operated automatically through Programmable Logic Controller (PLC) electrical apparatus control system. The integrated A/O MBR system was made from stainless steel 304 with a thickness of 2 mm. The total available volume of the bioreactor was 0.54 m^3 with 1.5 m in length, 0.6 m in width, and 0.6 m in water depth (0.9 m in total height). The integrated A/O bioreactor was divided into anoxic zone with available volume of 0.13 m^3 and aerobic zone with available volume of 0.41 m^3 by clapboard. Two hollow fibre microfiltration membrane modules made of Polyvinylidene fluoride (PVDF) were set in MBR system. The dimension of each membrane module was 0.30 m in length, 0.24 m in width and 0.25 m in height. The parameters of each membrane module are: membrane diameter 0.65 mm/1.00 mm (inside/outside); membrane pore size 0.2 μm; and membrane area 4.5 m^2.

1.3 Characterization index for membrane fouling

The expression of membrane flux is as Eq. (1):

$$J = \Delta p/(R_t \cdot \mu) \tag{1}$$

where, R_t (1/m) is the total membrane filtration resistance, μ (Pa·sec) is the sludge viscosity, and Δp (Pa) is the trans-membrane pressure.

During the investigation period, the membrane flux of treated water maintained at 40 L/hr through increasing the peristaltic pump power gradually. Since membrane flux (J) of water is constant, the membrane fouling can be characterized by the specific membrane flux (J_b) (Eq.(2)). That is, membrane fouling can be characterized by the change of trans-membrane pressure.

$$J_b = J/\Delta p = 1/(R \cdot \mu) \tag{2}$$

Equation (2) indicates main factors affecting membrane fouling are R and μ, which are inversely proportional to the specific membrane flux (J_b). Increases of R and μ may cause a decrease of J_b and a serious membrane fouling. The constitutes of R are mainly R_p and R_c since the changes of R_m can be ignored compared with R_p and R_c.

1.4 Analytical items and method

MLSS was determined by filtering the mixture liquor through a 1-μm membrane followed by drying the SS together with filtering membrane to a constant weight at 105°C (EPA, 2003). Sludge viscosity (μ) was determined by determination of oil viscosity at low temperature (GB/T506-82). Scanning electron microscope was obtained by HITACHI (Japan). Particle size was determined by laser grain size analysis.

Analytical method of extracellular polymeric substances (EPS) (Comte et al., 2006): to centrifuge a quantitative sludge mixture at 4000 r/min for 10 min to divide the sludge mixture into supernatant and sludge floc; to discard the supernatant and make up the volume with distilled water; to mix the sludge floc and the distilled water for 10 min followed by extracting for 20 min at 80°C water bath; to centrifuge the sludge mixture at 10000 r/min for 10 min after extraction, and to take the supernatant filtering through 0.45 μm membrane. EPS content can be characterized by the total organic carbon of the filtrate.

2 Results and discussion

2.1 Impact of MLSS and sludge viscosity on membrane fouling

In an MBR, R_c accounts for a large portion of R. The formation of sludge cake layer is directly related to MLSS. Figure 2 shows the relationship between MLSS and trans-membrance pressure (TMP) of two operation cycles (from Oct 3 to 11 and Oct 15 to 27) under similar returning flow of mixture, suction/suspended time and aeration intensity. A hydraulic cleaning of the membrane was conducted in Oct 13 during the intermittence of the two operation cycles.

Figure 2 shows that in the first operation cycle (total 9 days, see open symbols), MLSS concentration increases from around 3.5 to 5.0 g/L and the corresponding TMP increases from around 5×10^3 to 30×10^3 Pa. The increase rate of the TMP (or the slope of the linear growth trend) was approximately 3200 Pa/day. In the first 12 days of the second operation cycle (total 13 days, see solid symbols), MLSS concentration increases from around 5.0 to 7.8 g/L and the corresponding TMP increases from around 5×10^3 to 45×10^3 Pa. The increase rate of the TMP in the beginning 9 days of the second cycle was very close to that of the first cycle. After that, TMP had a sudden increase, and at day 13 of the second operation cycle, TMP increases significantly to around 67×10^3 Pa. The TMP of the investigated MBR system is not particularly correlated with the MLSS under the range of 3.0–8.0 g/L. For hyperhaline municipal wastewater treatment of the investigated

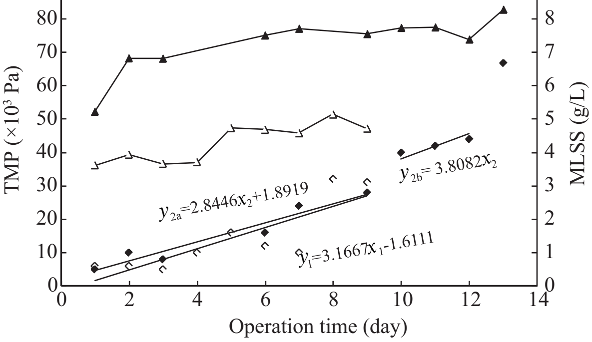

Fig. 2 Changes of trans-membrane pressure (TMP) and mixed liquor suspended solid (MLSS) against operation time. I, II mean first and second operation cycle, respectively.

MBR system, MLSS is not the main factor affecting trans-membrane pressure. Operation period is the main factor affecting the growth of trans-membrane pressure. Longer operation will lead to a rapid growth of trans-membrane pressure. Therefore, a regular membrane cleaning after around 9-day operation is necessary to ensure a reasonable operation cost and treated water quality.

Figure 3 shows the relationship between MLSS and μ during the operation of the MBR system from Sep 15 to 24 (total 10 days). The returning flow of mixture was 200 L/hr, the aeration intensity was 1.6 m^3/hr and the suction/suspended time was 12 min/3 min. During the operation period, the MLSS fluctuated in a range of 3.0 to 3.8 g/L and μ varies in a range of 2.88 to 3.30 Pa·sec. Variation trends of MLSS and μ against operation time are similar. A higher MLSS results in a greater μ. This is believed higher MLSS may produce more EPS, which leads to an increase in viscosity of the mixture. Considering the water treatment efficiency of MBR system and the stable operation of membrane modules, MLSS around 5.0 g/L is suggested and used in the following investigation of the hyperhaline municipal wastewater treatment.

2.2 Impact of microorganisms, extracellular polymeric substance (EPS) and sludge viscosity on membrane fouling

EPS is polymer material. It is produced by microorganisms under certain conditions and commonly found in interior and surface of activated sludge floc. In an MBR process, EPS may result in a stable gathering of microbial colony and a nice coordination of metabolism between populations. Although the adhesion of EPS can promote sludge granulation and increase sludge settling, it also causes the membrane fouling by absorption on the membrane surface (Zeng, 2007). Therefore, the changes of EPS concentration, sludge viscosity and TMP against operation time and their correlations were investigated. As shown in Fig. 4 during the investigation period, the returning flow of the mixture was approximately 200 L/hr, the aeration intensity was around 1.6 m^3/hr and the suction/suspended time was 12 min/3 min.

Figure 4 shows that during the investigation days, EPS content fluctuated in a range of 5.8 to 8.0 mg/g-MLVSS. Correspondingly, TMP is fluctuated in a range of 28×10^3 to 40×10^3 Pa and the sludge viscosity at around 2 Pa·sec. A relative high value of approximately 8.8 Pa·sec in the 2nd investigation day was believed to be that the dosage of urea in the last investigation period changes the growing environment of microorganisms. The overall fluctuation trends of TMP and sludge viscosity are well consist with EPS. Figure 4 shows a very good positive correlation among EPS, μ and TMP.

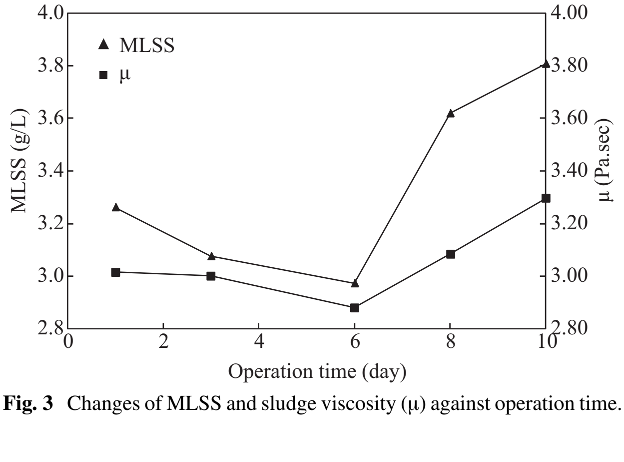

Fig. 3 Changes of MLSS and sludge viscosity (μ) against operation time.

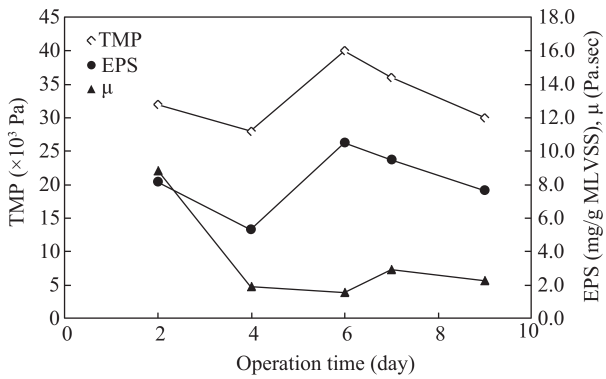

Fig. 4 Changes of extracellular polymeric substance (EPS), TMP and sludge viscosity against operation time.

To shed some light on the membrane fouling and the microbes of the mixture, a membrane thread and the mixture sludge of O zone were subjected to SEM (scanning electron microscope) analysis after investigation period (before membrane washing), as shown in Fig. 5.

Figure 5a shows that a large number of particles and floc attached onto the membrane surface and some particles inserted to the membrane pores. Figure 5b shows various microorganisms in the sludge, including large numbers of filamentous bacteria, long bacilli and short bacilli, etc. They staggered into frame, then adsorbed on the membrane surface or inserted into the membrane pores, causing the membrane pore size reduction. Thus, the trans-membrane pressure increased. Filamentous bacteria produce more EPS than floc bacteria (Chio et al., 2002). Microorganisms affect the viscosity through EPS producing, and further affect the membrane fouling.

2.3 Impact of aeration intensity on membrane fouling

Aeration has both positive and negative effects on membrane fouling, which depends on aeration intensity. Appropriate aeration intensity brings a proper fluid velocity and forms a proper hydraulic shear stress on the membrane surface. The cake layer deposited on the membrane surface may be blown off, the mixture of sludge can be air-washed and the mixture viscosity be reduced. Therefore, the membrane fouling will be reduced. However, too low or too high aeration intensity may cause serious membrane fouling. Too low aeration cannot produce enough shear force to blow off the cake layer. Too high aeration may cause destruction of the structure of activated sludge floc, the decrease of the sludge particle size and the release of EPS, which enhance the membrane fouling. Sludge particles close to the membrane pore sizes (approximately 0.2 μm in this study) most likely cause the membrane fouling (Lahoussine-Turcaud et al., 1990). Thus, effect of aeration intensity on TMP was investigated. The changes of TMP against investigation period under

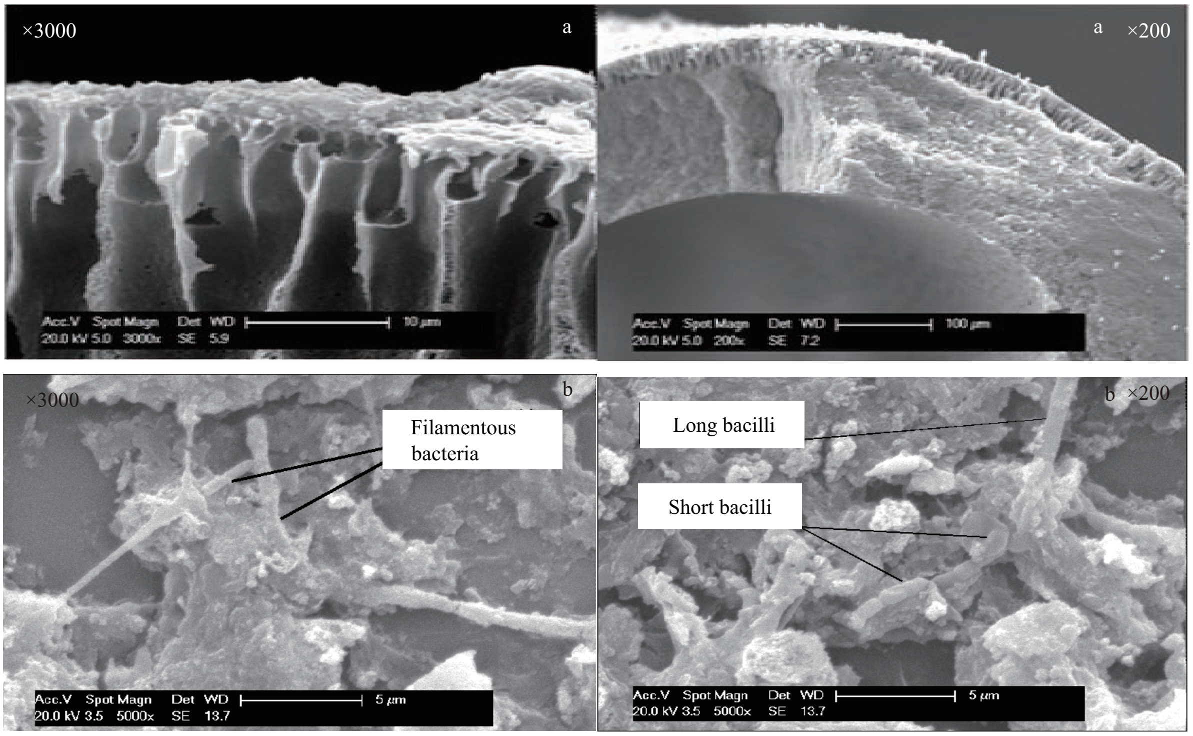

Fig. 5 Scanning electron microscopy (SEM) micrographs of a polluted membrane thread and the mixture sludge of O zone in A/O MBR system. (a) membrane wire cross-section scan before cleaning; (b) activated sludge micro.

three aeration intensities of 1.0, 1.6 and 1.8 m^3/hr are shown in Fig. 6. The operating conditions of the mixture returning flow and MLSS were in the same ranges for the three investigation periods.

Figure 6 shows that in the first 6 days, TMP rises rapidly from initial 20×10^3 to 28×10^3 Pa under the aeration intensity of 1.0 m^3/hr, from initial 28×10^3 to 36×10^3 Pa under the aeration intensity of 1.6 m^3/hr and from initial 5×10^3 to 54×10^3 Pa under the aeration intensity of 1.8 m^3/hr. The increasing rate of TMP or membrane resistance under the aeration intensity of 1.8 m^3/hr is much higher than those under the aeration intensity of 1.0 and 1.6 m^3/hr. The trend of TMP increase under the aeration intensity of 1.0 and 1.6 m^3/hr is similar. This indicates the diversion tubes under the membrane modules can strengthen the

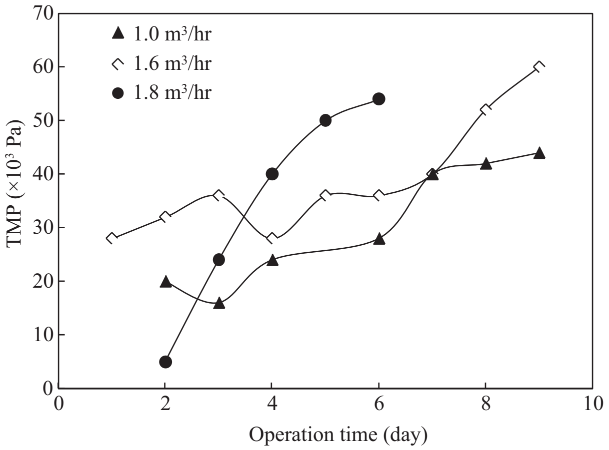

Fig. 6 Changes of TMP against operation time under different aeration intensity.

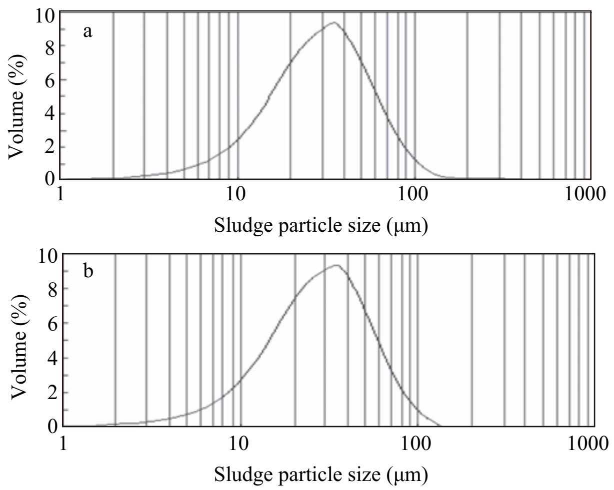

Fig. 7 Particle size distribution of sludge mixture. Under aeration intensity 1.0 m^3/hr (a) or 1.6 m^3/hr (b).

disturbance, enhance aeration efficiency and relieve the membrane pollution effectively. Therefore, the aeration intensity of 1.0 m^3/hr (air/water ratio of 25/1) is sufficient for the process.

Impact of aeration intensity on membrane fouling was also conducted by measuring the sludge diameter. Two aeration intensities of 1.0 and 1.6 m^3/hr were examined in the two investigation periods of Oct 18 to 26 and Nov 5 to 16, respectively, under MLSS of 5000–6000 mg/L. Sludge mixtures of O zone were withdrawn in Oct 25, and Nov 15 respectively. Particle size distributions of the samples were measured.

As shown in Fig. 7 under the aeration intensity of 1.0 m^3/hr, the average particle size of sludge is 35.56 μm and the average particle diameter of cumulating 10% volume, i.e. *D*(v, 0.1), is approximately 11.4 μm. Correspondingly, under the aeration intensity of 1.6 m^3/hr, the average particle size of sludge is 31.83 μm and the average particle diameter of cumulating 10% volume is approximately 10.7 μm. The sludge particle size under the aeration intensity of 1.6 m^3/hr is less than that of 1.0 m^3/hr. Increase of aeration intensity leads to the decrease of sludge particle size. However, almost all sludge particles were much greater than the average membrane pore size of 0.2 μm under the two aeration intensities. Thus, cake layer resistance should be the main membrane resistance and the main factor causing TMP increase for the treatment of hyperhaline municipal sewage.

Either of the two investigated aeration intensity cannot prevent the cake layer resistance. Moreover, increasing of aeration intensity has little or even negative effect on TN removal efficiency. Therefore, considering the economic costs, water treatment efficiency and the operation stability of the A/O MBR system, aeration intensity of 1.0 m^3/hr is suggested and used for the treatment of hyperhaline municipal sewage.

2.4 Impact of suction/suspended time on membrane fouling

Effect of suction/suspended time on TMP was investigated. The changes of TMP against investigation period under three suction/suspended time of 9 min/2 min, 12 min/3 min, 15 min/5 min are shown in Fig. 8. The operating conditions as the mixture returning flow, aeration intensity and MLSS were in the same ranges for the three investigation periods.

Figure 8 shows that in the first 6 days of each investigation period, TMP rises rapidly from initial 0.8×10^3 Pa to 54×10^3 Pa under the suction/suspended time of 15 min/5 min, from initial 10×10^3 to 31×10^3 Pa under the suction/suspended time of 9 min/2 min and from 4×10^3 to 18×10^3 Pa under the suction/suspended time of 12 min/3 min. The increasing rate of TMP or membrane resistance under the suction/suspended time of 12 min/3 min is much lower than those under the suction/suspended time of 9 min/2 min and 15 min/5 min.

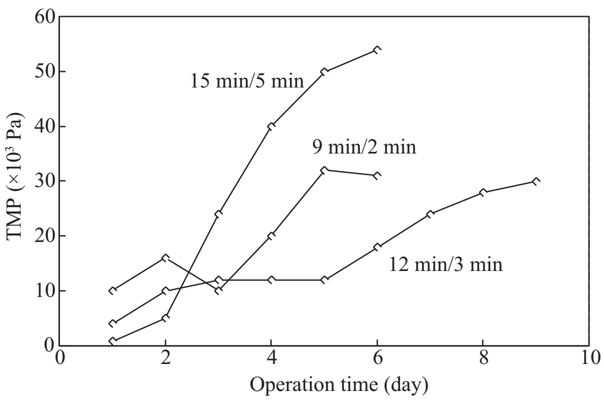

Fig. 8 Changes of TMP against operation time under different suction/suspended time.

Long suction time results in settling and accumulation of suspended solids and dissolved organic matters on membrane surface. Therefore, the pore blocking resistance (R_p) and the membrane surface cake layer resistance (R_c) increases. To maintain a constant treated water flux, TMP has to be increased. A proper suspended time is conducive to fall off the sludge particles from membrane surface due to the action of aeration and diffusion, similar results were reported by Gui et al. (2002) for the domestic wastewater treatment. However, too long suspended time cannot alleviate the increase of membrane resistance effectively. Instead, it decreases the water production. For a required water production, TMP increase may be effectively controlled by shortening the pump running time and increasing the frequency of pump stop. Therefore, the suction/suspended time of 12 min/3 min is suggested and used for the treatment of hyperhaline municipal sewage.

3 Conclusions

(1) High MLSS results in great concentration polarization. TMP is not particularly correlated with MLSS under 3.0–8.0 g/L for hyperhaline municipal sewage treatment by the investigated A/O-MBR system. MLSS is not the main factor affecting trans-membrane pressure while MLSS is lower than 8.0 g/L for hyperhaline municipal sewage treatment. However, considering the water treatment efficiency of the A/O-MBR system and the stable operation of the membrane modules, MLSS of 5 g/L is suggested for hyperhaline municipal sewage treatment.

(2) EPS, sludge viscosity (μ) and TMP have very good positive correlations. The numbers and species of microbial population affect the sludge viscosity through EPS producing, and further affect the membrane fouling.

(3) Either too low or too high aeration intensity may cause serious membrane fouling. Considering the economic costs, water treatment efficiency and the operation stability of the A/O-MBR system, aeration intensity of 1.0 m^3/hr is suggested for the treatment of hyperhaline municipal sewage.

(4) Long suction time results in settling and accumulation of suspended solids and dissolved organic matters on membrane surface. Long suspended time is conducive to fall off the sludge particles from membrane surface. However, too long suspended time cannot alleviate the increase of membrane resistance effectively. A suction/suspended time of 12 min/3 min is suggested for effective control of TMP increase for the treatment of hyperhaline municipal sewage.

(5) Various environmental factors and operation conditions have mutual influences. Properties of activated sludge mixture have direct impact on membrane fouling, and operation conditions have indirect impact on membrane fouling. There is a positive correlation among MLSS, EPS and sludge viscosity. Either numbers or species of microorganisms have impact on EPS concentration of the mixture. Intermittent suction of treated water can control the membrane fouling effectively under proper suction/suspended time and aeration intensity.

Acknowledgments

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