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Impact of Temperature Seasonal Change on Sludge Characteristics and Membrane Fouling in a Submerged Membrane Bioreactor

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Effects of temperature seasonal change on sludge characteristics and membrane fouling in a pilot-scale submerged membrane bioreactor (MBR) were studied. Results showed that bound extracellular polymeric substances (EPS), soluble chemical oxygen demand (SCOD), polysaccharides, and proteins in the supernatants all increased under low temperature operation. The increase of diluted sludge volume index (DSVI) and capillary suction time (CST) indicated that the settling and dewatering ability of mixed liquors deteriorated. It was also found that the increase of EPS, polysaccharides, and proteins led to the increase of biopolymers attached to membrane surfaces and in turn caused severe membrane fouling.

Keywords membrane bioreactor (MBR); membrane fouling; sludge characteristics; wastewater treatment

INTRODUCTION

Membrane bioreactor (MBR) is an innovative wastewater treatment technology, which combines an activated sludge process with membrane modules for solid-liquid separation. The MBR process has many advantages over a conventional activated sludge (CAS) system, such as a smaller footprint, less sludge production, and highly improved effluent quality (1,2). However, membrane fouling is still one of the main obstacles to the wide-spread application of this promising technology (3–5).

Many researchers have studied the effect of operation parameters and conditions on membrane fouling and filtration operation in submerged MBRs. Nuengjamnong et al. (6) investigated the influence of various sludge retention time (SRT) on extracellular polymeric substances (EPS) and on membrane fouling in submerged MBRs. They found that the extractable EPS was directly related to membrane fouling and the organic content of EPS

decreased as SRT increased. Similar results were reported by Grelier et al. (7). Drews et al. (8) found that unsteady states like intermittent feeding or shifts in the oxygen supply could lead to an increase in EPS formation or change in its fouling propensity. Ji and Zhou (9) observed that aeration intensity had an effect on microbial polymers and proteins/carbohydrates on membrane decreased with increasing aeration rates. The decrease of these organic polymeric substances on membranes explained why membrane fouling was reduced by increasing aeration rates. Rosenberger et al. (10) focused on the filtration characteristics of two parallel MBRs with only constructional difference (pre-denitrification and post-denitrification) and found that the non-settable fraction of the sludge (soluble and colloidal materials) was responsible for the different membrane fouling conditions between the two MBRs. Although these intensive efforts mentioned above are very helpful in understanding the relationship between different operating conditions and membrane fouling, there is a lack of information on the role of temperature seasonal change in the operation of submerged MBRs.

In this study, a pilot-scale MBR at an existing wastewater treatment plant (WWTP) was operated in order to identify the relationship between temperature seasonal change and sludge characteristics. The influence of temperature seasonal change on sludge characteristics, such as diluted sludge volume index (DSVI), capillary suction time (CST), soluble chemical oxygen demand (SCOD), and bio-polymer release (EPS, polysaccharides (PS), and proteins (PN)), etc. was investigated, and the effects on membrane fouling were also analyzed. The results obtained in this study are expected to provide a sound understanding of the role of temperature seasonal change in MBR operation.

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MATERIALS AND METHODS

Experimental Setup

The pilot-scale submerged membrane bioreactor as shown in Fig. 1, which was located in Quyang Municipal

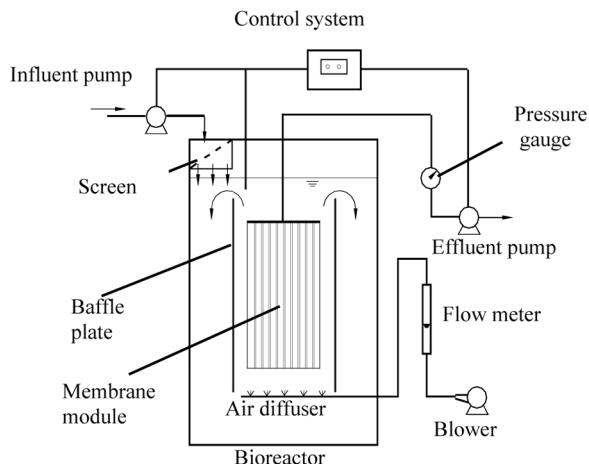


FIG. 1. Schematic flow diagram of the MBR.

WWTP of Shanghai, had a total volume of 1.44 m^3 ($0.8 \times 0.9 \times 2.0\text{ m}$ length \times width \times height). It was divided into a riser zone and two down-comer zones by two baffle plates. Thirty flat sheet membrane modules were located in the riser. The membranes were made of Polyether-sulfone (PES) membrane with a mean pore size of about $0.20\text{ }\mu\text{m}$. The effective filtration area for each module was 0.7 m^2 . Air was supplied through an air diffuser which was below the membrane modules in order to supply oxygen demanded by the microorganisms and induce a cross-flow velocity (CFV) along the membrane surface. After filtration through a stainless bar screen of 0.9 mm , raw wastewater from the WWTP was supplied into the bioreactor. The characteristics of the raw wastewater are listed in Table 1. The influent pump was controlled by a water level sensor to maintain a constant water level in the bioreactor over the experimental period. The membrane-filtered effluent was then obtained by suction using a pump connected to the modules. The effluent flow rate and the trans-membrane pressure (TMP) were monitored by a water meter and a pressure gauge, respectively.

Operating Conditions

The filtration operation of the pilot-scale MBR was conducted with the constant flow rate about $15\text{--}25\text{ L}/(\text{m}^2\text{ h})$,

TABLE 1
Characteristics of the raw municipal
wastewater (mg/L except pH)

Items	Concentration
COD	150–690
NH ₃ -N	17.8–58.2
TP	2.8–10.5
SS	60–500
pH	6.3–7.5

which was below the critical flux ($27\text{--}36\text{ L}/(\text{m}^2\text{ h})$). Intermittent filtration (10-min filtration and 2-min pause) was also carried out. The CFV along the membrane surfaces was maintained at $0.2\text{--}0.4\text{ m/s}$ by air scouring. The experiment was conducted at a hydraulic retention time (HRT) of $2.2\text{--}3.7\text{ h}$ (HRT was longer under low-temperature operation due to low flux operation; on the 440th day, seven membrane modules were broken and taken out which made HRT longer as well), a dissolved oxygen (DO) concentration about $1\text{--}2\text{ mg/L}$ in the bioreactor and sludge retention times (SRT) in the range of $10\text{--}40\text{ d}$. The TMP was in the range of $4\text{--}30\text{ kPa}$ and chemical cleaning-in-place procedure would be carried out if TMP was over 30 kPa .

Analytical Methods

Measurements of chemical oxygen demand (COD), total phosphorus (TP), ammonia (NH₃-N), and the pH in the influent and membrane effluent, mixed liquor suspended solids (MLSS), and mixed liquor volatile suspended solids (MLVSS) in the bioreactor were performed according to *Chinese NEPA Standard Methods* (11). The SCOD samples were obtained by filtrating the mixed liquor of the MBR through filter paper with a mean pore size $0.45\text{ }\mu\text{m}$. The DO concentration in the reactor was measured by a dissolved oxygen meter (Model YSI 58, YSI Research Incorporated, Ohio, USA). CFV was determined using a cup-type current meter (Model LS45A, Chongqing Hydrological Instrument Incorporated, Chongqing, China). Due to the high concentration of suspended solids in the submerged MBR, the sludge samples were diluted to about 4 g/L for sludge volume index (SVI) measurements to minimize the potential wall effects. The parameter obtained by this procedure was called DSVI. The measurement of proteins (PN) was carried out by Lowry methods (12). BSA was used as a standard. The phenol-sulfuric acid method of Dubois et al. (13) was used for polysaccharide (PS) determination. Glucose was used as a standard. The sludge sample taken from the MBR was centrifuged at 3200 rpm for 30 min . Then the remaining pellet was used to extract the bound EPS according to the thermal treatment method described by Chang and Lee (14). The EPS content was measured by analyzing volatile solids (VS) of these EPS solution obtained from above procedures. Dehydrogenase activity assay was according to the procedures described by Chen and Gu (15). The specific dehydrogenases activity (SDA) was expressed as enzyme activity per unit biomass per hour ($\mu\text{g-TF}/\text{mg-MLSS h}$). All the above analyses were conducted in duplicate, and their average values were reported.

Statistical analysis, including Pearson and Spearman's rank correlations, was carried out with the software of SPSS 11.0 produced by SPSS Incorporation (America) with the aim of characterizing the influence of sludge

characteristics (or temperature) on membrane fouling rate. The Pearson's product momentum correlation coefficient (r_p) was used to estimate linear estimations. The Pearson's coefficient r_p is always between -1 and $+1$, where -1 means a perfect negative correlation and $+1$ a perfect positive correlation while 0 means absence of relationship. Correlations are considered statistically significant at a 95% confidence interval ($P < 0.05$).

Fouling Rate Determination

The membrane fouling rate of mixed liquors in the biorreactor was determined using a mini-membrane module with a filtration area of 0.098 m^2 and using the same material and pore size of that used in the pilot-scale membrane bioreactor. The effective volume of the mini-MBR is 2.5 L . Mixed liquor samples were taken from the pilot-scale bioreactor at various operation times, and a filtration test of 30 min was conducted immediately using the mini-module. A similar short-term filtration operation was also carried out by other researchers to determine membrane fouling behavior (16). The filtration of the mini-membrane module was conducted with the constant flow rate about $45 \text{ L}/(\text{m}^2 \text{ h})$ while the change of TMP was monitored. The filtrate was returned to the mini-MBR in order to keep constant characters of the mixed liquor. The CFV about 0.2 m/s along membrane surface was kept during the filtration run. The degree of membrane fouling was evaluated by membrane filtration resistance calculated by the following equation:

$$J = \frac{\text{TMP}}{\mu R} \quad (1)$$

where J is the membrane permeate flux ($\text{m}^3/(\text{m}^2 \text{ s})$), TMP is the trans-membrane pressure (Pa), μ is the permeate water viscosity (Pa s), and R is the membrane filtration resistance ($1/\text{m}$).

The membrane fouling rate (ΔR) of the mixed liquor was finally computed as $\Delta R_{30}(1/(\text{m h}))$, the change rate of R within 30-min filtration, according to Eq. (2).

$$\Delta R_{30} = \frac{R_0 - R_{30}}{\Delta t} \quad (2)$$

where Δt is filtration time (h), R_0 and R_{30} are the R values of the mini-module at starting time and after 30-min filtration, respectively ($1/\text{m}$).

By combining Eqs. (1) and (2), ΔR_{30} can be rewritten as Eq. (3):

$$\Delta R_{30} = \frac{\text{TMP}_{30} - \text{TMP}_0}{\mu J \Delta t} \quad (3)$$

where TMP_0 and TMP_{30} are the TMP values of the mini-module at starting time and after 30-min filtration,

respectively (Pa). In order to eliminate the influence of water viscosity variations on filtration resistance, the parameters μ and J used in Eq. (3) were adjusted to 20°C equivalent values.

Quantification the Contribution of Three Sludge Fractions to Membrane Fouling

Sludge fractions were separated according to Bouhabila's method (17). Three samples were available for the filtration tests—the initial sample of sludge (microbial flocs + colloids + solutes); a sample containing the colloids and solutes; and a sample containing only the solutes. The filtration tests were conducted using the mini-MBR as mentioned above but with 6-hour filtration. The various membrane resistances were computed by Eq. (1).

All membrane modules were soaked in hypochlorite sodium solution (v/v 0.5%) and washed with pure water several times to remove impurities on the membrane surface before filtration experiments. R_m was obtained by filtration of clean water and computed by Eq. (1), membrane resistance of three samples (i.e., the initial sample of sludge, a sample containing the colloids and solutes and a sample containing only the solutes) was named as R_{t1} , R_{t2} , R_{t3} , respectively. Then the resistance of microbial flocs, colloids, and solutes can be obtained, which was $(R_{t1} - R_{t2})$, $(R_{t2} - R_{t3})$ and $(R_{t3} - R_m)$, respectively.

RESULTS AND DISCUSSION

Process Performance

Figure 2 presents the variations of the membrane flux and trans-membrane pressure (TMP) during over 2-year operation. It could be observed that at lower temperature the TMP increased more rapidly and the flux also tended to decline even when operated at constant flux operation mode. In order to further understand this phenomenon, a series of analysis of sludge property change and membrane fouling characteristics during temperature seasonal change (take the period of day 45–day 180 as an example, temperature varying from 26°C to 8°C , SRT 40 d, and HRT 2.2 h (HRT was longer under low temperature operation)) was carried out.

The treated water quality is listed in Table 2. It can be seen that the removal of COD, ammonia and suspended solids was quite successful. Over 60% total phosphorus removal was also achieved during the performance. The phosphorus removal was mainly due to two factors—one was the biological utilization, the other the membrane separation function owing to the fine membrane pores (18).

Sludge Concentration Variations in the MBR

Sludge concentration variations during the experiment are shown in Fig. 3. From day 1 to day 44, at SRT 10 d and 24–26°C of mixed liquor, the growth of biomass in

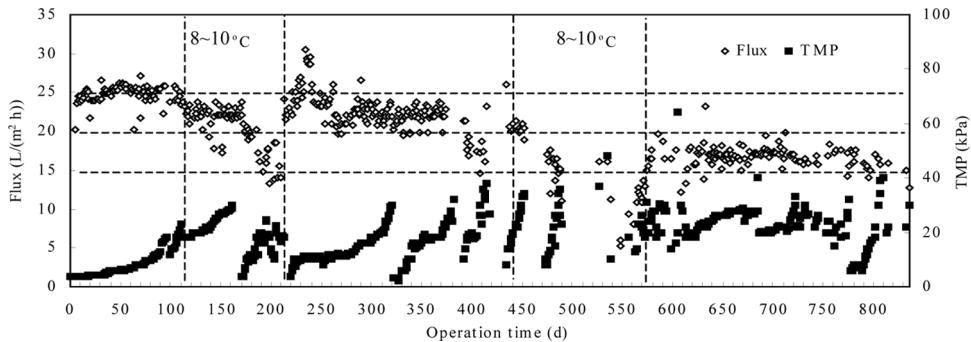


FIG. 2. Variations of membrane flux and TMP with operation time.

the MBR occurred after seeding the sludge and feeding wastewater, and a steady state with MLSS concentration 10–11 g/L was observed later. The biomass started to grow again from day 45 to day 148 when SRT 40 days was performed. A steady state of sludge concentration about 22 g/L was achieved at the end of this period. The decline of sludge biomass was observed from day 149 to day 181 when the temperature of the mixed liquor was in a range of 8–10°C. It was mainly attributed to the decline of volumetric organic loading (VOL) and the decrease of the sludge yield coefficient at such low temperature. From day 182 to day 301, as the temperature increased, MLSS began to grow and at last levelled off at 25 g/L. Then MLSS started to decrease when SRT was changed to 20 days from day 302 to day 380, and finally a steady state with MLSS concentration 14 g/L was reached. After day 380, MLSS tended to increase again. On the 440th day, seven broken membranes were taken out from the reactor and caused a low VOL which made MLSS decrease. The MLSS concentration decreased in the period of day 500 to day 600 because of low temperature and increased after day 600 due to the increase of temperature.

From day 45 to day 181, the biomass characteristics in the MBR including DSVI, Bound EPS, SCOD, CST, PN, and PS, etc., were systematically measured in order to verify the interaction of temperature (temperature gradually decreased from 26°C to 8°C in the MBR) to the sludge properties and membrane fouling.

TABLE 2
Quality of treated water (mg/L)

Items	Concentration
COD	10–38
NH ₃ -N	0.1–7.8
TP	1.1–3.2
SS	0

Variations of Sludge Characteristics

From Fig. 4, it can be seen that DSVI increased as the temperature declined from 26°C to 8°C, which indicated that the sludge settling ability deteriorated. The decrease of temperature also resulted in the increase of SCOD of the supernatant, polysaccharides, and proteins in the supernatant, and EPS concentration of the mixed liquor. The higher biopolymer concentration in the MBR at lower temperature might be explained by a higher production of soluble microbial products and by a reduced degradation of these substances due to kinetic considerations (19). The variation of the dewatering ability in terms of CST values in this period was also studied (see Fig. 5), and the results showed that CST increased rapidly with the decline of temperature. The statistical analysis by using SPSS, as listed in Table 3, demonstrated that temperature has negative correlations with EPS, CST, SCOD, polysaccharides, proteins, MLSS, and the membrane fouling rate.

Polymers Deposited on Membrane Surfaces

In order to classify the effect of different sludge properties on the membrane fouling, a membrane module with an effective filtration area 0.098 m² was successively submerged in the pilot-scale MBR from day 97 to day 102 and from day 151 to day 156. The membrane constant flux of this membrane module was conducted at 30 L/(m²·h). On day 102 and day 156, the membrane foulants were collected from the membrane surface, respectively. Then the PS, PN, and the EPS composition of the membrane foulants were further analyzed. The test results are listed in Table 4.

From Table 4, it can be found that the quantity of the membrane foulants attached to the membrane surface from day 151 to day 156 is more significant than that from day 97 to day 102. It also indicates that polymeric substances in the mixed liquor have certain correlations with membrane foulants deposited on the membrane surface. The more the polymeric substances existed in mixed liquors, the more the membrane foulants deposited on the membrane surfaces.

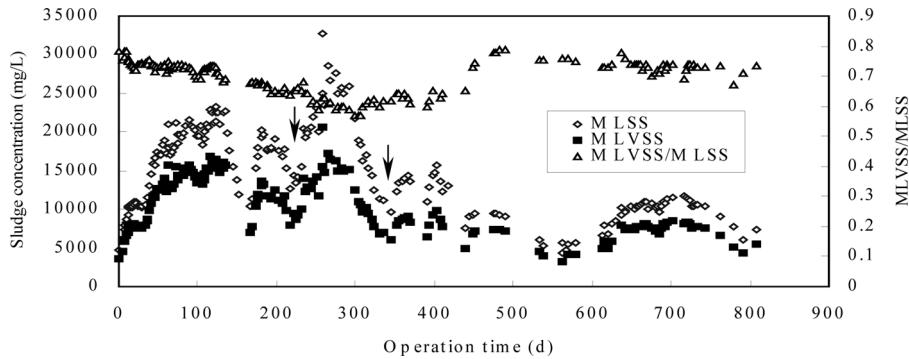


FIG. 3. Variations of sludge concentration during the whole performance.

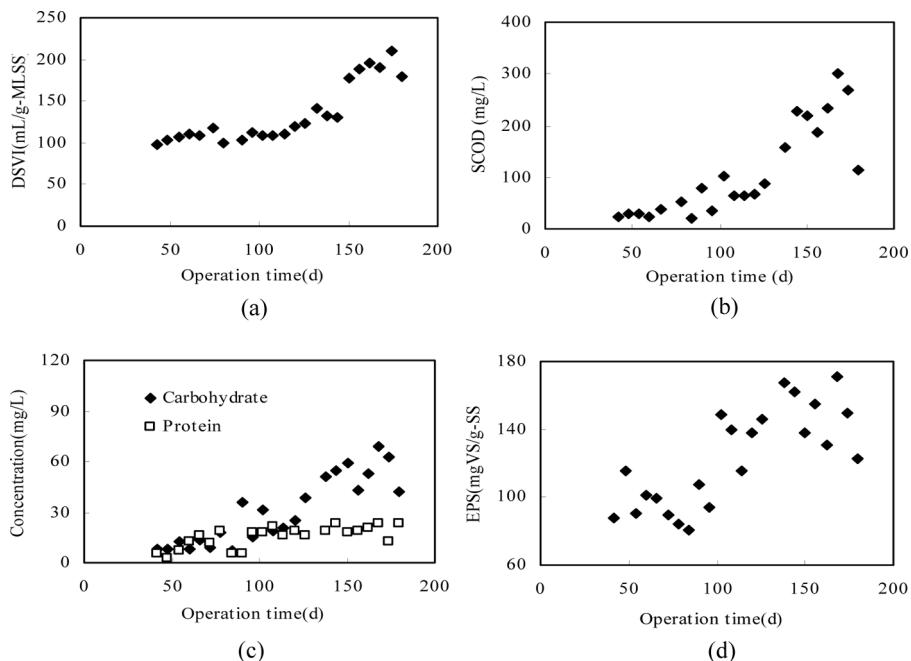


FIG. 4. Variations of (a) DSVI, (b) soluble COD, (c) polysaccharides and proteins in supernatant, and (d) EPS during temperature seasonal change from day 45 to day 180.

From day 97 to day 102, the TMP of this membrane module increased from 2.5 kPa to 10.5 kPa; however, the TMP from day 151 to day 156 increased from 2.4 kPa to 30 kPa. It also showed that the membrane fouling occurred more dramatically due to the membrane foulant deposition on the membrane surface or membrane pore blocking from day 151 to day 156 compared with that from day 97 to day 102.

Relationship Between Sludge Characteristics and Membrane Fouling

From day 45 to day 181, the membrane fouling rate of the mixed liquors in terms of ΔR_{30} was determined. The correlations between the membrane fouling rate and the

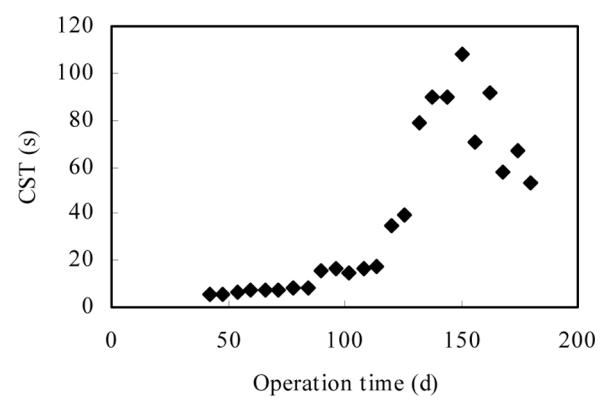


FIG. 5. CST variations during temperature seasonal change period.

TABLE 3
Pearson correlations' coefficient (r_p) of various parameters and fouling rate

Items	MLSS	EPS	CST	SCOD	PS	PN	ΔR_{30}
Temperature	-0.111	-0.781**	-0.771**	-0.740**	-0.733*	-0.516**	-0.720**

**Correlation is significant at 0.01 level (2-tailed).

TABLE 4
Quantification of membrane foulants attached to membrane surfaces (mg/m²-membrane)

No.	Operation period	PS	PN	EPS ^a
1	Day 97–102	82.0	54.0	136.0
2	Day 151–156	262.5	214.5	477.0

^aEPS are considered as the total amount of carbohydrates and proteins.

variables of sludge characteristics were further analyzed according to the statistical analysis method. Test results show that the correlation coefficient r_p of DSVI, bound EPS, PS, PN, SCOD with membrane fouling rate is 0.873 ($P < 0.01$), 0.762 ($P < 0.01$), 0.891 ($P < 0.01$), 0.538 ($P < 0.05$), 0.896 ($P < 0.01$), respectively. It can be seen that the variations of sludge characteristics have significant impacts on the membrane fouling behavior. The statistical analysis also shows that the temperature has a strong negative effect on the membrane fouling rate (the correlation coefficient $r_p = -0.870$ ($P < 0.01$)). It showed that membrane fouling rate increased as the temperature decreased.

In order to further verify the effect of sludge properties on membrane fouling, the contribution of sludge compositions to membrane fouling was investigated on day 97 and day 151 (temperature gradually decreased from 26°C to 8°C in the MBR), respectively. The result is shown in Table 5.

It can be seen from Table 5 that the total resistance on day 151 is higher than that on day 97. The contribution of microbial flocs to membrane resistance on day 97 and day 151 was almost the same; however, the resistance caused by colloids and solutes on day 151 was much higher

than that on day 97. On day 97 and day 151, the MLSS concentration in the MBR was 20.8 g/L and 21.5 g/L, respectively. It indicates that the effect of different MLSS concentration on membrane fouling between day 97 and day 151 can almost be neglected. The main reason that contributed to the increase of membrane resistance was the increase of colloids and solutes in the MBR at low temperature. It confirmed that a low operation temperature had a significant effect on sludge properties and membrane fouling.

Analysis of the Effect of Temperature on Sludge Characteristics

The microbial activities in terms of SDA were measured during the operation. From day 45 to day 148, the average SDA was 5.5 µg-TF/(mg-MLSS h) ($n = 10$); however, the average SDA dropped to 3.3 µg-TF/(mg-MLSS h) ($n = 5$) during the run from day 149 to day 181. It was evident that the microbial activities were lowered at low temperature in the MBR.

From the analysis of the above sections, it is also known that the increase of microbial polymeric substances (mainly classified into EPS, or soluble microbial products (SMP) including PS and PN, etc.) is a significant factor contributing to severe membrane fouling at low temperature. A common theme of EPS and SMP is that they are microbially produced organic materials that contain electrons and carbon, but are not active cells (20). The diversion of electrons and carbon affects cell yield and sludge growth rate. The traditional view is that all electron-donor oxygen demand (OD) is either shunted to the electron acceptor to generate energy or is converted to biomass (21). In fact, a part of OD is shunted to EPS or SMP formation and the OD available for synthesizing active biomass is reduced.

TABLE 5
Contribution of various fractions in sludge to membrane fouling^a

Fractions	Day 97	Day 151
Microbial flocs (1/m)	1.72×10^{12} (66.9%)	1.52×10^{12} (32.0%)
Colloids (1/m)	4.50×10^{11} (17.5%)	1.69×10^{12} (35.6%)
Solutes (1/m)	4.01×10^{11} (15.6%)	1.54×10^{12} (32.4%)
Total resistance (1/m)	2.57×10^{12} (100%)	4.75×10^{12} (100%)

^aThe data in parentheses are the relative contribution percentage of various fractions to membrane fouling.

As shown in Fig. 3, from day 149 to day 181, MLSS (MLVSS) concentration in MBR declined. It demonstrated that the active biomass yield and specific growth rate declined even operated at almost the same volumetric organic load (VOL) as before. That well indicated that part of OD was shunted to EPS or SMP formation in the MBR during this period. The results shown in Fig. 4 also confirm that the EPS and SMP in the MBR increased significantly from day 149 to day 180.

According to the unified theory developed by Laspidou and Rittmann (20), cells use electrons from the electron-donor substrate to build active biomass, and they also produce bound EPS and utilization-associated products (UAP) at the same time and in proportion to substrate utilization. Bound EPS are hydrolyzed to biomass-associated products (BAP), while biomass undergoes endogenous decay to form residual dead cells. They verified that soluble EPS is actually SMP, or the sum of UAP and BAP, which are biodegradable and utilized by active biomass as recycled electron-donors substrate. As discussed above, microbial activities in terms of SDA at low temperature were lowered. Therefore, the degradation of soluble EPS (or SMP) was reduced. In the MBR, the microbial polymeric substances are mainly dependent on the balance of their formation and hydrolysis (dissolution). The formation and hydrolysis of polymeric substances were kept relatively balanced and the polymeric substances in the MBR did not vary significantly during the smooth operation; however, when MBR was operated at low temperature, as analyzed above, the balance was broken. Large quantities of EPS were produced by microorganisms while their EPS hydrolysis ability was lowered. That resulted in accumulation and increase of microbial polymeric substances in MBR during the operation at low temperature.

CONCLUSIONS

The effect of temperature seasonal change on sludge characteristics and membrane fouling was studied in a pilot-scale submerged membrane bioreactor (MBR) for real municipal wastewater treatment, and the following conclusions could be drawn.

Bound EPS extracted from biomass, SCOD, PS, and PN in the supernatants all increased obviously under low temperature operation. The increase of EPS, PS, and PN in the MBR led to the increase of biopolymers attached to the membrane surface and in turn caused severe membrane fouling during low temperature operation in MBRs.

It was also found that DSVI and CST increased as the temperature decreased, which indicated the settling ability and dewatering ability of mixed liquors deteriorated.

Temperature seasonal change had significant impacts on the microbial physiology and could increase a higher

production of biopolymers and a reduced degradation ability of these substances.

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