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Mitigated membrane fouling in a vertical submerged membrane bioreactor (VSMBR)

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Abstract

In a laboratory-scale study, characteristics of membrane fouling in an A/O (anoxic/oxic) series membrane bioreactor (MBR) and in a vertical submerged membrane bioreactor (VSMBR) treating synthetic wastewater were compared under the same operating conditions. Accordingly, fouling characteristics of a pilot-scale VSMBR treating municipal wastewater were studied under various operating conditions. Various physical, chemical, and biological factors were used to describe membrane resistances. As a result, it was concluded that high concentrations of extracellular polymeric substances (EPS), high viscosity and a high sludge volume index (SVI) corresponded to high membrane resistance indicating severe membrane fouling in both the laboratory-scale MBRs and the pilot-scale VSMBR. In addition, high fouling potential was observed in the pilot-scale VSMBR at 60-day sludge retention time (SRT). In this case, as hydraulic retention time (HRT) decreased from 10 to 4 h, EPS concentrations increased and the average particle size increased, leading to reduced settling of the sludge and increased membrane fouling. To mitigate fouling, two different methods using air bubble jets were adopted in the pilot-scale VSMBR. As a result, it was found that air backwashing was more efficient for fouling mitigation than was air scouring.

Keywords: Vertical submerged membrane bioreactor; Membrane fouling; EPS; Particle size; Air bubble jets

1. Introduction

Because nutrients such as phosphorus and nitrogen in municipal wastewater have been recognized as major culprits contributing to eutrophication the effluent quality standards at wastewater treatment plants are becoming more stringent. During the activated sludge process, treated water is stored in a large tank where the biomass separates from the effluent by settling. The effluent quality is limited by the difficulty of separating the suspended biomass. In addition, the process generates large quantities of sludge for which treatment and disposal represent 50% of total treatment cost [1]. One viable alternative for the activated sludge processes is use of membrane bioreactors (MBRs) that maintain a high biomass concentration in the reactor. This approach, which has the potential to reduce hydraulic retention time (HRT) and excess sludge while permitting a higher rate of organic matter removal is currently attracting a great deal of attention from academia and industry [2,3].

Before the membrane bioreactor is fully ready for field application, however, some of its limitations must be addressed. First of all, there is often a rapid decline in flux due to membrane fouling as a result of the high biomass concentration in the reactor. Membrane fouling is characterized as a reduction of permeate flux through the membrane resulting from increased membrane resistance due to pore blocking, concentration polarization, and cake formation [4]. Permeate flux decline is influenced by a number of factors relating to the feed wastewater (composition), the membrane (element geometry/configuration, area and material composition), and reactor operation (biological condition and hydrodynamics). As characteristics of the membrane are determined by membrane manufacturers, researchers have focused on various strategies to reduce membrane fouling. These strategies include development of a new design of membrane module [4,5] and modification of the feed flow pattern [6].

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Particle sizes of the pollutants in wastewater may strongly affect fouling mechanisms in a membrane filtration system. If foulants are comparable, or smaller than the membrane pores, adsorption and pore blocking may occur. However, if foulants are much larger than the membrane pores, they tend to form a cake layer on the membrane surface. For a microfiltration membrane process for treating wastewater, biofouling is a major problem because most foulants (microbial flocs) in MBRs are much larger than the membrane pore size. Biofouling may start with the deposition of individual bacteria on the membrane surface, after which the cells multiply and form a cake layer. Many researchers suggest that soluble microbial product (SMP) and extracellular polymeric substances (EPS) secreted by bacteria also play important roles in formation of biological foulants and cake layer on membrane surfaces [7,8].

Many researchers investigating SMP fouling by refractory organic matter in activated sludge [9,10], have found that the higher the SMP concentration is, the faster the membrane fouling proceeds [11]. Accordingly, Kim et al. [12] and Nagaoka et al. [7] reported a decrease in filterability with increasing suspended and extractable EPS concentrations, respectively. EPS are composed of many organic compounds such as polysaccharides, amino polysaccharides and proteins [13] and are considered to aid in floc formation and enhance microbial attachment to membrane surfaces, preventing detachment by mechanically cross-linking and stabilizing the biofilm.

In the previous work [14], a new vertical submerged MBR (VSMBR) design including anoxic (lower layer) and oxic zones (upper layer) in one reactor was fabricated and operated under various conditions. Due to efficiencies related to its physical structure, this design is expected to challenge current limits to the effective removal of nutrients from wastewater as well as for the reduction of excess sludge [14,15]. The objective of this study was to compare characteristics of membrane fouling between a laboratory-scale series MBR and a laboratory-scale VSMBR. In addition, effects of various operating conditions such as different HRTs and total chemical oxygen demand to total nitrogen (T-COD/T-N) ratios on membrane fouling were studied in the pilot-scale VSMBR fed with several different organic substrates.

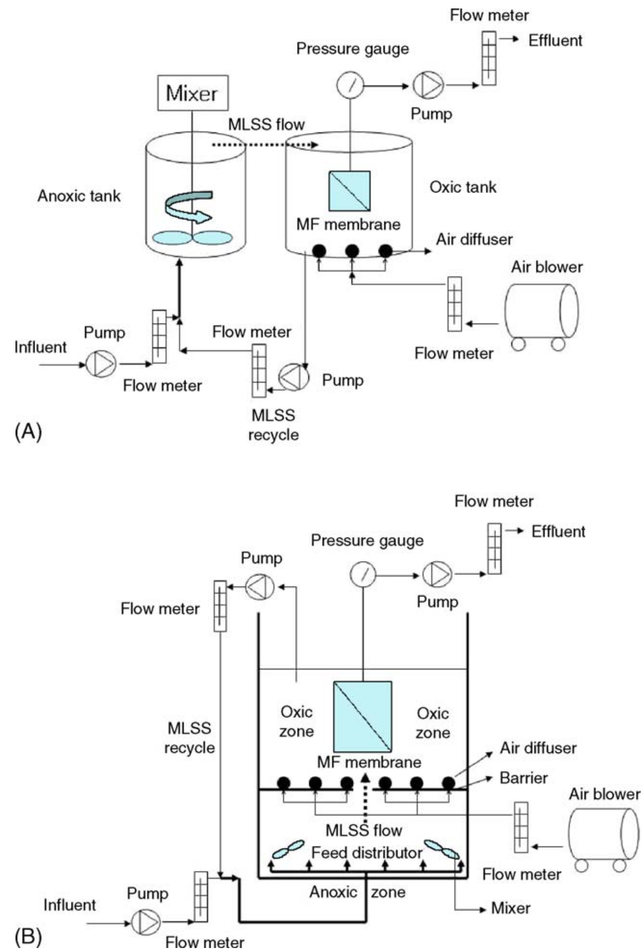

Fig. 1. Schematic diagrams of an A/O series MBR (A) and a VSMBR (B).

2. Material and methods

2.1. Experimental set-up for two laboratory-scale MBRs fed with synthetic wastewater

The VSMBR that includes both anoxic and oxic zones in one reactor was developed as shown in Fig. 1, in an attempt to remove organic matter and nutrients effectively, while reducing membrane fouling and subsequent operation cost as well. To study the effect of physical characteristics of an MBR on membrane fouling, two MBRs with different configurations were operated. As shown in Fig. 1, the MBRs both contained anoxic and oxic zones.

In the VSMBR, influent was introduced into the anoxic zone (lower layer). In the anoxic zone, the supplied organic matter was first used for denitrification and phosphorus release. Complete mixing in the anoxic zone was obtained by using a low speed mixer (30–50 rpm). A mixed liquid of suspended solids (MLSS) flows from the anoxic zone (bottom) to the oxic zone (upper layer) through a hole (diameter of 10 cm) in the center. The aerobic zone was separated from the anoxic zone by a horizontal plate. As a result, this flow from the anoxic zone (lower layer) to the oxic zone (upper layer) resulted in higher concentrations of MLSS in the anoxic zone by gravity settling of microorganism than in the oxic zone. In the aerobic zone, disk type diffusers were used to provide air bubbles for oxidation of organics and ammonia, and to reduce membrane fouling.

In the case of the A/O series MBR, the influent was also introduced into the anoxic zone, but there was no vertical flow from the bottom to the top. Thus that MLSS moved in uniform concentration from the anoxic into the oxic zone. And complete mixing in the anoxic zone was obtained by using a constant speed mixer (about 150 rpm). As a result, MLSS concentrations within the anoxic and oxic zones were similar.

Both MBRs were inoculated with sludge taken from a municipal wastewater treatment plant in Daejeon, Korea. The pH, and volatile suspended solid to total suspended solid (VSS/TSS) ratio of the sludge were 7.2 and 0.73, respectively. Effluent was withdrawn from the oxic zones of the two MBR systems through

Table 1
Characteristics of the PTFE membrane

Category	Characteristic
Material	Poly-tetrafluoroethylene
Pore size (μm)	0.45
Module type	Hollow fiber
Outer/inner diameter (mm)	2/1
Maximum temperature (°C)	95
Operation range of pH	0–14

Table 2
Characteristics of the synthetic wastewater for the two laboratory-scale MBRs

Item	Compound	Concentration (mg l^{-1})
Organic	Glucose	160 as COD
Nitrogen	Ammonium sulfate	40 as N
Phosphorus	Potassium phosphate	6 as P
Alkalinity	Sodium bicarbonate	200 as $CaCO_3$
Trace elements	Calcium chloride	0.50
	Cobalt chloride	0.35
	Cupric sulfate	0.15
	Ferric chloride anhydrous	0.80
	Magnesium sulfate	0.34
	Manganese chloride	0.50
	Sodium molybdate dihydrate	0.20
	Yeast extract	10
	Zinc sulfate	0.55

a microfiltration (MF) membrane made of PTFE (Sumitomo Electric Fine Polymer Inc., Japan) having a nominal pore size of 0.45 μm (Table 1).

To avoid any fluctuation in the influent, and to provide a stable source of completely biodegradable organics, synthetic wastewater having total T-COD concentration of 160 mg l^{-1}, T-N concentration of 40 mg l^{-1}, and total phosphorus (T-P) concentration of 6 mg l^{-1} with trace nutrients was supplied to the MBRs from a storage tank. Sodium bicarbonate (200 mg l^{-1} as $CaCO_3$) was added to the influent of both MBRs to maintain a constant pH and to support nitrification. Details of the composition of the influent are summarized in Table 2.

As presented in Table 3, two laboratory-scale MBRs having different configurations were operated continuously to study the effect of configuration on characteristics of membrane fouling under similar conditions. Average MLSS concentrations in the anoxic and the oxic zones of the VSMBR were about 5.7 and 2.2 g l^{-1}, respectively. However, in the A/O series MBR, the average MLSS concentrations were about 4.5 g l^{-1} in both zones.

Table 3
Operating conditions of the two laboratory-scale MBRs

Parameter	A/O series MBR		VSMBR	
	Anoxic zone	Oxic zone	Anoxic zone	Oxic zone
SRT (days)	30		30	
HRT (h)	3	5	3	5
Working volume (l)	6	10	12	20
Permeate flux ($l\,m^{-2}\,h^{-1}$)	6.2 (=0.15 m day^{-1})			
Operation mode	On (8 min)/idle (2 min)		On (8 min)/idle (2 min)	
Internal recycle rate (%)	400	–	400	–
ORP (mV)	−63	+314	−147	+257

Table 4
Operating conditions of the pilot-scale VSMBR

Parameter	Pilot-scale VSMBR	
	Anoxic zone	Oxic zone
SRT (days)	60	
Working volume (l)	500	833
Average MLSS (g l^{-1})	8.68	4.21
ORP (mV)	−153	+274

2.2. *Experimental set-up for a pilot-scale VSMBR treating municipal wastewater*

Table 4 describes operating conditions of a pilot-scale VSMBR that was continuously operated under the same conditions as the two laboratory-scale MBRs except for influent qualities for variety and SRT of 60 days. Characteristics of the municipal wastewater and the external carbon source used are shown in Table 5. The total COD/T-N ratio of the sewage was found to be about 5.5 indicating too little carbon for complete nutrient removal. In order to increase the nutrient removal efficiency, organic-rich liquid substrates in the form of condensate of food waste (CFW) were added. The CFW was produced from food waste in a high-rate fermenting drier operated at 110 °C and HRT of 12 h. The CFW contained high concentrations of VFAs (1% lactic acid, 79% acetic acid, 11% propionic acid, and 9% butyric acid).

HRT was considered a key factor among various operating parameters because removal efficiencies of nitrogen and phosphorus in the biological nutrient removal (BNR) process were

Table 5
Characteristics of municipal wastewater and the external carbon source for the pilot-scale VSMBR

Constituents	Municipal wastewater	CFW
COD (mg l^{-1})		
Total	232 ± 41	13750 ± 170
Soluble	161 ± 25	13200 ± 260
SS (mg l^{-1})		
Total	220 ± 52	Not detected
Volatile	110 ± 30	Not detected
Nitrogen (mg l^{-1})		
Total	42 ± 5	83 ± 26
NH_3–N	27 ± 3	55 ± 18
NO_3–N	Not detected	Not detected
Phosphorus (mg l^{-1})		
Total	3.2 ± 0.4	36 ± 15
Soluble	1.9 ± 0.4	15 ± 9
VFAs as COD (mg l^{-1})	Not detected	5100 ± 58
Initial pH	7.3 ± 0.1	4.0 ± 0.1
Alkalinity as $CaCO_3$ (mg l^{-1})	125 ± 47	–

Table 6
Experimental schedule for the pilot-scale VSMBR

Phase	HRT (h)	Total COD/T-N ratio	Operation period (days)	Remark
1	10	5.5	29–68	Without CFW
2	8	5.5	69–360	Without CFW
3	6	5.5	361–480	Without CFW
4	4	5.5	481–600	Without CFW
5	8	6.8	601–630	With CFW (0.43%, vol./vol.)
6	8	8.2	631–660	With CFW (0.86%, vol./vol.)

most dependent on HRT. Changes in HRT indicated variation of hydraulic loading rates for the organics, and other nutrients, affecting membrane fouling. Therefore, as shown in Table 6, the effects of various HRTs (Phases 1–4) on membrane fouling were studied in the pilot-scale VSMBR. In BNR processes, nutrient removal efficiencies are very sensitive to both the quantity and characteristics (especially biodegradability) of organic sources, because poly-P accumulating organisms (PAOs) and denitrifiers require organic matter for phosphorus release and denitrification [16]. Because most domestic wastewater in Korea has insufficient organic content for effective nutrient removal, the effect on membrane fouling from using CFW as a supplementary carbon source to improve nutrient removal efficiency, was also studied in Phases 5 and 6.

2.3. Mitigating membrane fouling using air bubble jets

Because higher TMP leads to greater membrane fouling, in submerged MBRs the permeate flux is relatively low, resulting in extended periods without a severe decline in flow. However, at some time during the filtration process, permeate flux declines to a level where it is no longer economical to continue and the filter must be cleaned. A number of cleaning options exist, with backwashing and/or chemical cleaning being the most common. However, chemicals such as sodium hypochlorite solution or nitric acid for in-situ cleaning can be toxic to microorganisms. In response to this concern, two air supply techniques were developed and tested to remediate fouling. As shown in Fig. 2, in one method (i.e. air scouring) bubbles from air nozzles were directed onto the exterior membrane surface. In the other method (i.e. air backwashing) air bubbles were injected from inside through the hollow fiber module. The permeate flux and air flow rates of the two air bubble jets techniques were maintained at 18.6 l m^{-2} h^{-1} and 300 l m^{-2} s^{-1}, respectively. For this study, effluent of the two MBR systems was withdrawn for 8 min (suction pump and aeration on; air bubbles for backwashing off) and air bubbles for backwashing supplied for 2 min (suction pump and aeration off).

2.4. Membrane resistance analysis model

The resistance-in-series model was used to analyze membrane fouling resistances, which describes the permeate flux–TMP relationship over the entire domain of pressure [17].

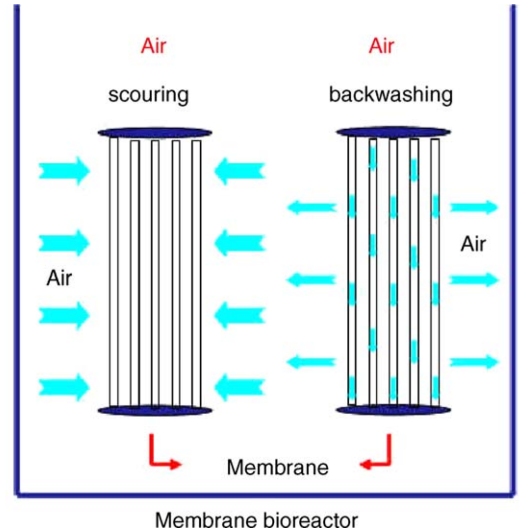

Fig. 2. Schematic diagrams of two fouling mitigation methods using air bubble jets.

Based on this model, the permeate flux on the applied TMP can be described by Darcy's law as Eq. (1):

$$J_v = \frac{1}{A}\frac{dV}{dt} = \frac{\Delta P}{\mu R_t} = \frac{\Delta P}{\mu(R_m + R_c + R_{a+p})} \quad (1)$$

where J_v is the permeate flux ($m^3\, m^{-2}\, s^{-1}$), V the total volume of permeate (m^3), A the membrane area (m^2), ΔP the TMP (Pa), and μ is the dynamic viscosity of permeate (Pa s). Total membrane resistance R_t (m^{-1}) was composed of intrinsic membrane resistance R_m (m^{-1}), adsorption and pore blocking resistances R_{a+p} (m^{-1}), and cake layer resistance R_c (m^{-1}).

To test the fouling characteristics of the samples, a stirred cell having an effective volume of 200 ml (Amicon, USA) was used with the YM 30 membrane (MWCO of 30 kDa, i.d. = 62 mm, effective membrane surface area = 28.7 cm^{-2}, Millipore, USA). As shown in Fig. 3, the filtration protocol was similar to that employed by Taniguchi et al. [18]. All membrane modules were soaked and washed with deionized (DI) water several times to

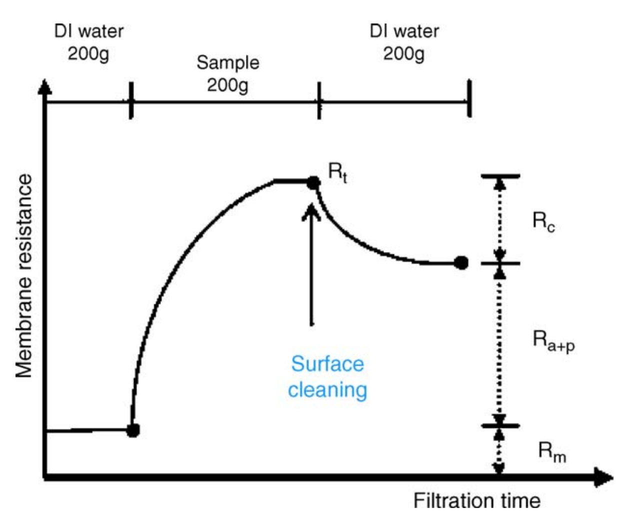

Fig. 3. Filtration protocol for the membrane resistance analysis.

remove impurities on the membrane surface before filtration experiments. Each membrane was compacted at 80 kPa. After that the pressure was decreased to an operational pressure of 20 kPa, and the intrinsic membrane resistance was measured. R_t was obtained by filtration of the sludge sample. For R_{a+p}, the cake layer was first removed then permeate flow was measured. R_c was estimated from R_t and R_{a+p} values. To avoid temperature effects, the resistances were normalized to 25 °C.

2.5. Determination of various membrane fouling parameters

In general, the membrane fouling occurring in membrane bioreactors is attributable to three processes: sludge particle deposition, adhesion of macromolecules to the membrane surface, and pore clogging by small molecules [19]. In this study, particle size of microbial flocs, hydrophobicity, and concentrations of EPS and SMP were selected as physical, chemical, and biological fouling factors, respectively. The floc size distribution of the mixed liquor was measured by a particle size analyzer having a range of 0.5–500 μm (Model 770, Particle Sizing Systems, USA). In this device, the scattered light reaches a detector that converts the signal to a size distribution based on volume. The average size of the flocs was given as the mean based on the volume equivalent diameter. Specific ultraviolet absorbance (SUVA) was used to estimate the hydrophobicity of the supernatant. SUVA is the ratio of UV absorbance at 254 nm (UV_{254}) to dissolved organic carbon (DOC) concentration. High SUVA indicates high degree of hydrophobicity due to a relatively large aromatic component.

One of the major biological fouling factors, SMP is composed of a wide range of organic compounds that differ in structure and molecular weight. SMPs have been found to include humic and fulvic acids, polysaccharide, proteins, nucleic acid, organic acids, amino acids, antibiotics, steroids, enzymes, structural components of cells, and products of energy metabolism [20]. The removal of biodegradable organic matter (BOM) represents the consumption of the influent substrate that should be equal to the removal of DOC if no SMP were present. If SMP were present, the DOC removal would be reduced by the amount of SMP produced. The concentration of produced SMP is the same as DOC in the effluent because BOM is completely removed when completely biodegradable organic substrates, such as acetic acid, propionic acid and glucose, are provided as a substrate. If it can be assumed that the produced SMP was not biodegraded in the reactor, then the amount of SMP produced in the reactor can be simplified as equivalent to DOC concentration in the effluent. In practice, the SMP concentration can be estimated using the BOM removal and the DOC removal as shown in Eq. (2):

$$\mathrm{SMP} = (\mathrm{BOM_{in}} - \mathrm{BOM_{out}}) - (\mathrm{DOC_{in}} - \mathrm{DOC_{out}}) + \mathrm{SMP_{biod}} \quad (2)$$

where SMP is the produced soluble microbial product in the reactor (mg l^{-1}), BOM_{in} the biodegradable organic matter in the influent (mg l^{-1}), BOM_{out} the biodegradable organic matter in the effluent (mg l^{-1}), DOC_{in} the dissolved organic carbon in the influent (mg l^{-1}), DOC_{out} the dissolved organic carbon in the effluent (mg l^{-1}), and SMP_{biod} is the biodegradable soluble microbial product in the reactor (mg l^{-1}).

In contrast, EPS was extracted from the microbial floc using a heat treatment [17]. In this study, the EPS was classified as proteins and carbohydrates, which are the dominant components typically found in EPS [21]. To determine protein EPS, the method proposed by Lowry et al. [22], modified by Frølund et al. [23] was used with a bovine serum albumin (BSA) as a standard material. Carbohydrate EPS was determined according to the phenol-sulfuric acid method with glucose as a standard material [24]. Viscosity of the mixed liquor was measured using a rotational viscometer (Model LVDV, Brookfield, England) at 20 °C. Due to the non-Newtonian nature of the sludge flow, the viscosity depends on a shear rate gradient determined by pressure and temperature. The viscosity, expressed as mPa s (centi-poise, cP), was measured at the shear rate of 100 s^{-1} for 5 min that the entire measurement would occur while the sludge was still in suspension. The SVI is defined in Eq. (3):

$$\mathrm{SVI} = \frac{H_{30} D}{H_0 C_0} \quad (3)$$

where SVI is the sludge volume index (ml g^{-1}), H_0 initial sludge height (cm), H_{30} the sludge height after 30 min settling (cm), C_0 the initial sludge concentration (g ml^{-1}), and D is the dilution factor.

One liter samples from the anoxic and the oxic zones in the MBRs were analyzed for various fouling parameters every other day during the experimental period. Oxidation–reduction potential (ORP) was measured using ORP electrodes connected to a pH meter (Orion Model 420A, Orion Research Inc., USA).

3. Results and discussion

3.1. Characteristics of membrane fouling in the two laboratory-scale MBRs having different configurations

To study characteristics of membrane fouling, two laboratory-scale MBRs having different configurations of anoxic and oxic zones were operated under the same conditions (HRT, SRT, and internal recycle rate). Fig. 4 shows variations of permeate flux and TMP of the anoxic/oxic series MBR and the VSMBR. As shown in those figures, in the initial step, the TMP increased to only 4 kPa up to 20 days and then increased gradually to 30 kPa, indicating a decrease of permeate flow rate up to 59 days in the A/O series MBR. In contrast, the TMP was steady at 2 kPa up to 20 days and then took 94 days to increase gradually to 30 kPa in the VSMBR.

After each experiment, the mixed liquors of the A/O series MBR and the VSMBRs were sampled to compare the fouling factors in both MBRs. To compare the process performance of the MBRs, specific removal rates (SRR, kg COD or T-N or T-P m^{-3} day^{-1}) of organic matter, T-N, and T-P were selected. The SRRs of COD, T-N, and T-P were calculated according to Eq. (4). Data obtained from the comparison of removal efficiencies of various substances and fouling factors obtained in this study

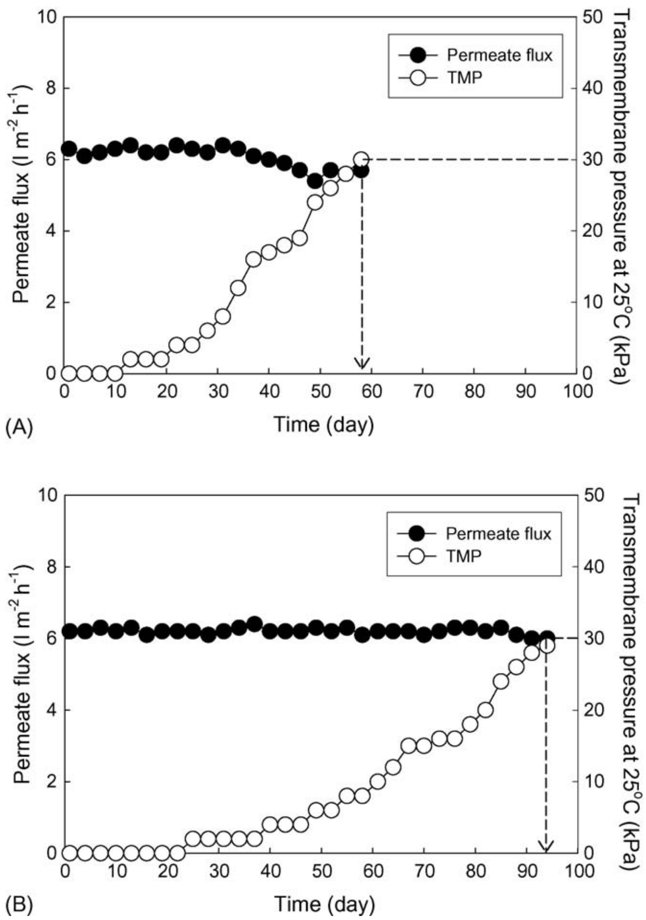

Fig. 4. Variations of flux and TMP in the A/O series MBR (A) and the VSMBR (B).

were given in Tables 7 and 8, respectively.

$$SRR = \frac{(C_{in} - C_{out})Q}{WV \times 1000} \quad (4)$$

where C_{in} is the concentration of COD or T-P or T-P in the influent (mg l^{-1}), C_{out} the concentration of COD or T-P or T-P in the effluent (mg l^{-1}), Q the treatment capacity (l day^{-1}), WV is the working volume of the reactor (l).

As shown in Table 7, the laboratory-scale VSMBR did better than the A/O series MBR in terms of removal efficiencies and SRR values of organic matter, T-N, and T-P. This might be caused by the vertical structure of the VSMBR. Due to its design, organic matter was used for the denitrification and phosphorus release by a relatively high concentration of microorganisms in the anoxic zone, and only a small amount of organic matter was passed into the oxic zone. This resulted in high nitrification performance and low excess sludge [14]. Moreover, as shown in Table 8, a comparison between the two laboratory-scale MBRs, revealed that the A/O series MBR showed more severe fouling. This could have resulted from relatively higher values of EPS contents per VSS and viscosity, and relatively smaller average particle size that the VSMBR.

3.2. *Characteristics of membrane fouling in the laboratory-scale and pilot-scale VSMBRs*

Based on the results of the laboratory-scale study, a pilot-scale VSMBR treating municipal wastewater was operated at the long SRT of 60 days for about 21 months under various operating conditions [15]. As shown in Fig. 5, it took 58 days to reach the TMP of 30 kPa in the pilot-scale VSMBR at 8 h-HRT. As shown in Table 7, a comparison between the laboratory-scale and the pilot-scale VSMBRs indicated that there was not much difference in terms of removal efficiencies of organic matter, T-N, and T-P. However, as shown in Table 8, the membrane resistance by cake layer formation was about four times higher than that of the laboratory-scale VSMBR at the same HRT of 8 h, due probably to the increased EPS contents, viscosity, hydrophobicity, and SVI value (indicating the settleability of sludge). In addition, it was found that the average particle size of the sludge was smaller resulting in increased membrane resistance due to adsorption and pore blocking in the pilot-scale VSMBR. Those differences in membrane fouling between the VSMBRs at different scales might have been caused by unfavorable conditions such as long SRT (60 days), low F/M ratio, and

Table 7
Comparison of removal efficiencies of organic matter, T-N, and T-P between the A/O series MBR and the VSMBRs

Item	A/O series MBR	VSMBR	
		Laboratory-scale	Pilot-scale [15]
Influent	Synthetic wastewater (glucose)		Municipal wastewater
HRT (h)	8	8	8
SRT (days)	30	30	60
Internal recycle rate (%)	400	400	400
F/M ratio (day^{-1})[a]	0.11	0.14	0.12
Temperature (°C)	20.5 ± 1.0	21.0 ± 1.5	19.3 ± 1.0
Overall T-COD removal efficiency (%)	96	95	96
SRR of T-COD in the reactor (kg COD m^{-3} day^{-1})	0.461	0.456	0.665
Overall T-N removal efficiency (%)	70	78	74
SRR of T-N in the reactor (kg N m^{-3} day^{-1})	0.084	0.094	0.093
Overall T-P removal efficiency (%)	63	81	78
SRR of T-P in the reactor (kg P m^{-3} day^{-1})	0.011	0.015	0.007

[a] F/M ratio (kg T-COD/kg MLSS day^{-1}).

Table 8
Comparison of various fouling factors for the A/O series MBR and the VSMBRs

Item	A/O series MBR	VSMBR	
		Laboratory-scale	Pilot-scale
Operation period to 30 kPa (days)	59	94	58
Particle size (vol. wt. mean, μm)	90.5 ± 2.5	98.4 ± 1.0	55.6 ± 4.5
Hydrophobicity ($l\,m^{-1}\,mg^{-1}$)	0.008 ± 0.001	0.006 ± 0.001	0.014 ± 0.003
SMP as DOC ($mg\,l^{-1}$)	3.4 ± 0.3	4.7 ± 0.2	3.1 ± 0.2
EPS carbohydrates ($mg\,g\,VSS^{-1}$)	48.4 ± 1.4	34.2 ± 3.7	64.9 ± 2.1
EPS proteins ($mg\,g\,VSS^{-1}$)	35.1 ± 1.1	32.8 ± 0.9	85.6 ± 8.3
Viscosity (mPa s)	1.55 ± 0.10	1.31 ± 0.05	1.45 ± 0.02
SVI ($ml\,g^{-1}$)	247 ± 15	279 ± 8	315 ± 20
Membrane resistance			
R_t ($\times 10^{12}\,m^{-1}$)	5.5 ± 0.2	3.4 ± 0.1	13.5 ± 0.4
R_m ($\times 10^{12}\,m^{-1}$)	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1
R_{a+p} ($\times 10^{12}\,m^{-1}$)	1.9 ± 0.2	1.2 ± 0.1	3.9 ± 0.5
R_c ($\times 10^{12}\,m^{-1}$)	3.4 ± 0.4	2.0 ± 0.1	9.4 ± 0.7

influent characteristics (municipal wastewater having a high SS concentration).

3.3. Fouling characteristics in the pilot-scale VSMBR at various HRTs

There are several limitations to wider use of the MBR process for domestic wastewater treatment. One of the important limitations has been fouling problems and subsequent increase of long-term operating costs. In these regards, to optimize operating conditions with reduced membrane fouling, the relationship between fouling factors and membrane resistance was studied. Fig. 6(A) and (B) show the relationships between carbohydrate EPS content per VSS, protein EPS content per VSS and various membrane resistances.

As shown in those figures, membrane resistance increased as HRT decreased from 10 to 4 h. In addition, carbohydrate EPS concentration increased from 23 to 140 $mg\,l^{-1}$ when HRT decreased from 10 to 4 h. However, protein EPS concentration increased only about 30 $mg\,l^{-1}$ under the same operating condition. As a result, it was concluded that the total membrane resistance was strongly affected by cake layer formation resis-

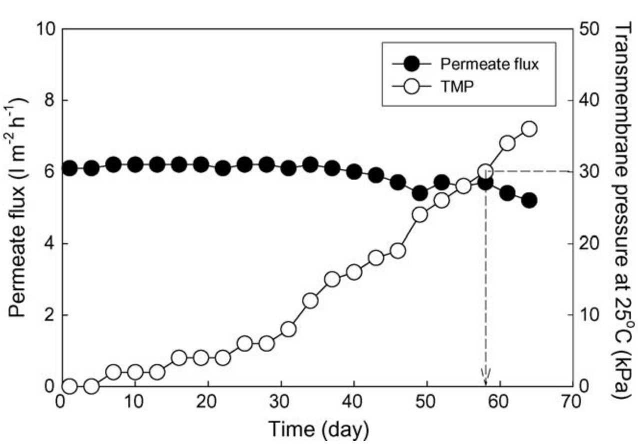

Fig. 5. Variations of flux and TMP in the pilot-scale VSMBR at 8-h HRT.

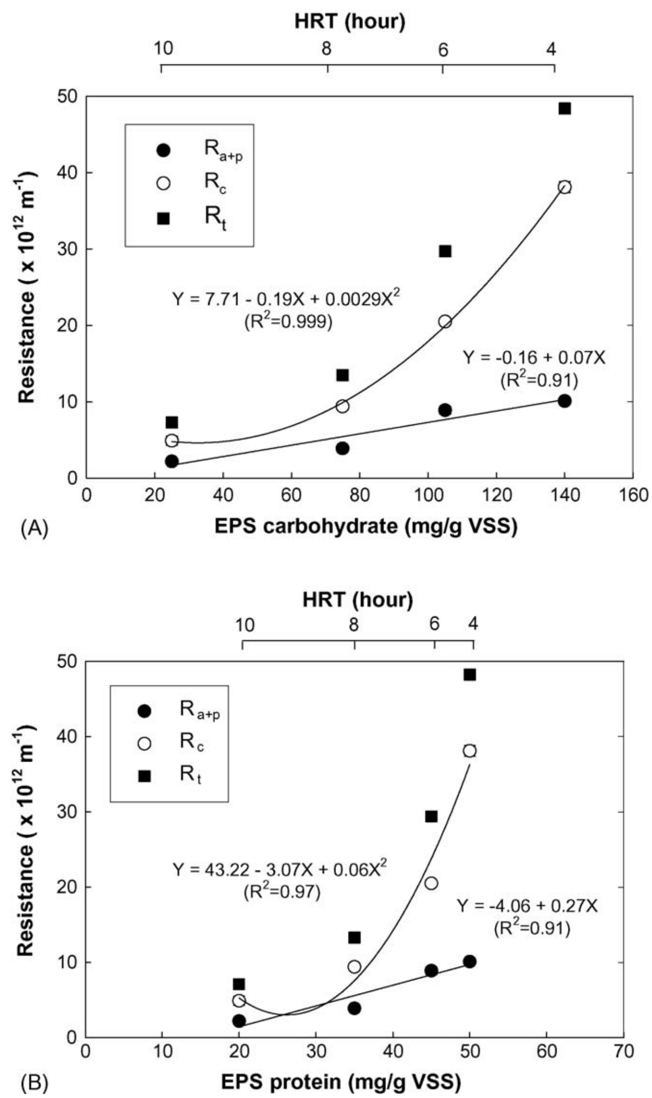

Fig. 6. Effects of EPS carbohydrate (A) and EPS protein (B) on fouling in the pilot-scale MBR at various HRTs.

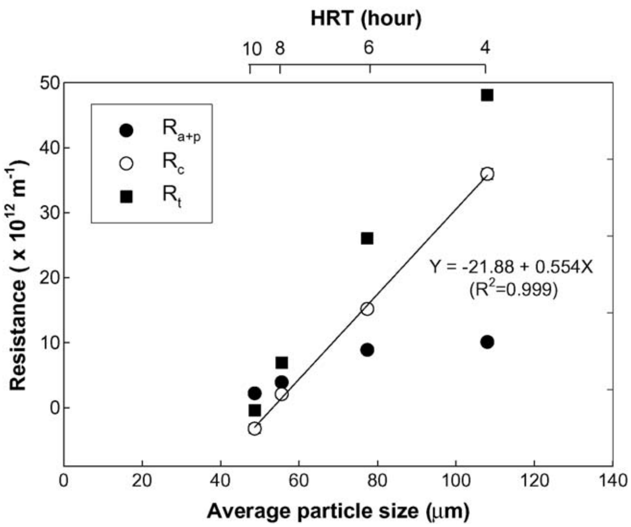

Fig. 7. Effects of particle size on membrane fouling in the pilot-scale VSMBR at various HRTs.

tance, and that this resulted from carbohydrate EPS rather than protein EPS.

On the other hand, the cake layer resistance was proportional to the average pore size of the microbial flocs. However, as shown in Fig. 7, the effect of average pore size of microbial flocs on the adsorption and pore blocking resistance was limited to those within a range of 45–80 μm. According to the Carman–Kozeny equation for cake filtration, water permeability is mainly influenced by particle size and cake porosity. Permeability increases dramatically as porosity and particle size increase [25]. Based on this equation, the most important parameter does appear to be porosity because particle size enters as a square, whereas porosity is cubic.

It has been shown that higher EPS concentration can cause poor cake layer permeability [26] and that EPS might also cause an increase in viscosity of the mixed liquor, as well as an increase in filtration resistance of the membrane [7]. It may be that the excretion of EPS is the most important promoter of flocculation in the mixed liquor [27], since the EPS are believed to enhance the biosorption of particles.

When others investigated the role of the bacterial extracellular matrix in resistance and solute rejection, their results showed that EPS and cells co-deposit on the membrane surface, filling the voids between the cells [28]. This co-deposition could cause formation of a potentially compressible cake which, due to its high hydraulic resistance, could result in the reduction of its porosity. In this study, as HRT decreased from 10 to 4 h, carbohydrate and protein EPS contents increased (Fig. 6). For the reasons, discussed above, this combination may have caused the observed increase of particle size, the increase in viscosity of the mixed liquor and the decrease of cake porosity resulting in the severe fouling of the membrane.

As mentioned in the previous study [15], supplementation of the external carbon source (CFW) improved nutrient removal efficiency from municipal wastewater having a low total COD/T-N ratio. However, as shown in Table 9, the supplementation with CFW also caused an adverse effect on membrane fouling

Table 9
Characteristics of membrane resistance in the pilot-scale VSMBR at various HRTs

Phase	Operating condition	Resistance ($\times 10^{12}$ m^{-1})			
		R_m	R_{a+p}	R_c	R_t
1	HRT 10 h	0.2	2.2	4.9	7.3
2	HRT 8 h	0.2	3.9	9.4	13.5
3	HRT 6 h	0.3	8.9	20.5	29.7
4	HRT 4 h	0.2	10.1	38.1	48.4
5	HRT 8 h (addition of CFW 0.43%)	0.2	4.1	10.1	14.4
6	HRT 8 h (addition of CFW 0.86%)	0.2	4.0	13.4	17.6

indicated by the fact that the total membrane resistance was higher by 7% and 30% in Phases 5 and 6, respectively, than was the total resistance in Phase 2 at the same 8-h HRT.

3.4. Fouling mitigation in the pilot-scale VSMBR using air bubble jets

To mitigate membrane fouling in the pilot-scale VSMBR, two kinds of fouling mitigation techniques using air bubble jets were introduced. As shown in Fig. 4, in the initial step, the TMP was

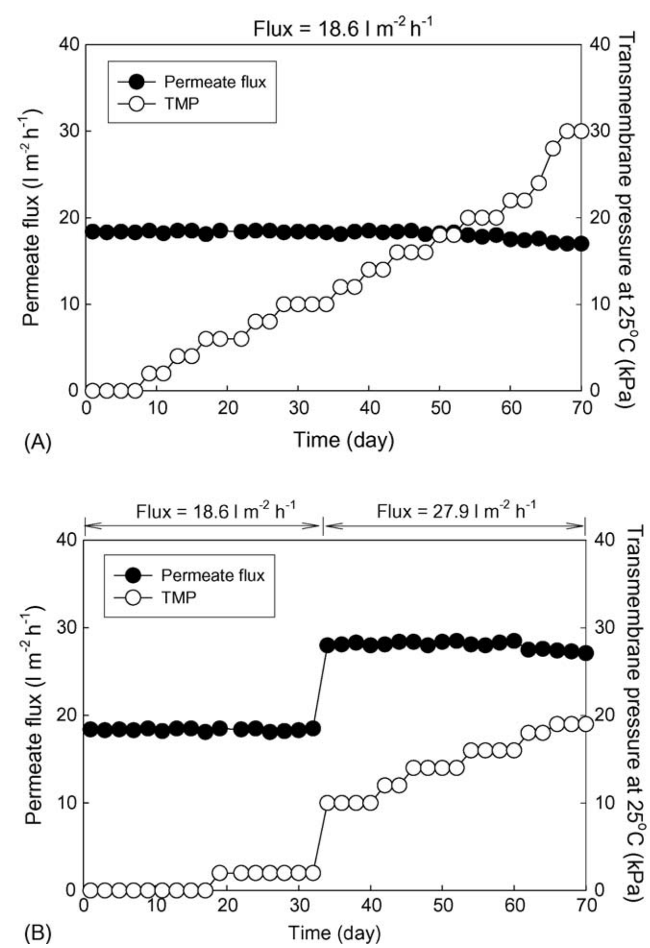

Fig. 8. Fouling mitigation by air scouring (A) and air backwashing (B) in the pilot-scale VSMBR.

maintained below 4 kPa until 10 days and then increased gradually to 30 kPa showing the decrease of permeate flow rate at 70 days. As a result, the pilot-scale VSMBR was operated without chemical cleaning for 70 days when the exterior membrane mitigation method was applied regardless of the high permeate flow rate of 18.61 $l\,m^{-2}\,h^{-1}$. However, in the case of the MBR using air backwashing, the TMP was maintained below 4 kPa until 30 days at the permeate flux of 18.61 $l\,m^{-2}\,h^{-1}$ (Fig. 8).

The permeate flux increased from 18.6 to 27.91 $l\,m^{-2}\,h^{-1}$ at 32 days. After that, the TMP was gradually increased and maintained below 20 kPa to the 70th days. From these experiments, it was concluded that both fouling mitigation techniques were effective for extending the operating period without chemical cleaning, and that the interior injection method (i.e. air backwashing) was more efficient than the exterior method (i.e. air scouring). It appeared that interior injection removed not only the cake layer, but also the adsorbed foulants blocking the pores. In contrast, the exterior injection appeared only to remove the cake layer from the membrane surface.

4. Conclusions

In this study, characteristics of membrane fouling in the laboratory-scale A/O series MBR and the VSMBR treating synthetic wastewater were investigated under the same operating conditions (HRT, SRT, and internal recycle rate). In addition, fouling characteristics of a pilot-scale VSMBR treating municipal wastewater was also studied under various operating conditions. Physical (average particle size of the microbial flocs), chemical (hydrophobicity), and biological factors (EPS contents and viscosity) were used to describe membrane resistances in this work. As a result, the following conclusions were drawn:

1. The A/O series MBR exhibited a greater fouling tendency than the VSMBR due to relatively higher values of EPS contents and viscosity, and relatively smaller average particle size than that of the VSMBR. In these regards, the VSMBR showed more potential for reducing membrane fouling.
2. The resistance due to cake layer formation was about 60% of total resistance in all MBRs. High values of protein and carbohydrate EPS concentration, viscosity and SVI corresponded to high membrane resistance indicating severe membrane fouling in both the A/O series MBR and the VSMBR treating synthetic wastewater.
3. A comparison between the laboratory-scale and the pilot-scale VSMBRs revealed that high fouling potential was observed in the pilot-scale VSMBR. It was operated relatively longer SRT and at a lower F/M ratio than the laboratory-scale VSMBR. An explanation for the difference in filtration performance was found by comparing fouling factors. As a result, it was concluded that as HRT decreased from 10 to 4 h, EPS concentrations and the average particle size increased, deteriorating sludge settleability (viscosity and SVI) and membrane resistances. Therefore, HRT would be considered an important operating parameter to reduce membrane fouling.
4. Pore blocking and adsorption, and cake layer resistance played important roles in microfiltration membrane fouling. Among them, the cake layer resistance (R_c) appeared to be the controlling factor of the total resistance in any MBR and under any operating conditions.
5. Two fouling mitigation techniques using air bubble jets enabled operation at high filtrate fluxes without severe membrane fouling in the pilot-scale VSMBR. The interior injection method (i.e. air backwashing) was more efficient than the exterior method (i.e. air scouring). It appeared that interior injection removed not only the cake layer, but also the adsorbed foulants blocking the pores. As a result, it was possible to prolong active flux, rather than having to clean the membranes completely. In contrast, the exterior injection appeared only to remove the cake layer from the membrane surface.

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Nomenclature

A	membrane surface area (m^2)
BOM_{in}	biodegradable organic matter in the influent ($mg\,l^{-1}$)
BOM_{out}	biodegradable organic matter in the effluent ($mg\,l^{-1}$)
C_{in}	concentration of COD or T-N or T-P in the influent ($mg\,l^{-1}$)
C_{out}	concentration of COD or T-N or T-P in the effluent ($mg\,l^{-1}$)
C_0	initial sludge concentration ($g\,ml^{-1}$)
D	dilution factor
DOC_{in}	dissolved organic carbon in the influent ($mg\,l^{-1}$)
DOC_{out}	dissolved organic carbon in the effluent ($mg\,l^{-1}$)
H_0	initial sludge height (cm)
H_{30}	sludge height after 30 min settling (cm)
J_v	permeate flux ($m^3\,m^{-2}\,s^{-1}$)
ΔP	transmembrane pressure (Pa)
Q	treatment capacity ($l\,day^{-1}$)
R_{a+p}	adsorption and pore blocking resistances (m^{-1})
R_c	cake layer resistance (m^{-1})
R_m	intrinsic membrane resistance (m^{-1})
R_t	total membrane resistance (m^{-1})
SMP	produced soluble microbial product in the reactor ($mg\,l^{-1}$)
SMP_{biod}	biodegradable soluble microbial product in the reactor ($mg\,l^{-1}$)
SRR	specific removal rate of COD or T-N or T-P in the influent ($kg\,m^{-3}\,day^{-1}$)

SVI sludge volume index (ml g^{-1})
V total volume of permeate (m^3)
WV working volume of the reactor (l)

Greek symbol
μ dynamic viscosity of permeate (Pa s)

References

[1] L. Defrance, M.Y. Jaffrin, Comparison between filtrations at fixed transmembrane pressure and fixed permeate flux: application to a membrane bioreactor used for wastewater treatment, J. Membr. Sci. 152 (1999) 203–210.
[2] K.H. Krauth, K.F. Staab, Pressurised bioreactor with membrane filtration for wastewater treatment, Water Res. 27 (3) (1993) 405–411.
[3] C. Chiemchaisri, K. Yamamoto, Performance of membrane separation bioreactor at various temperatures for domestic wastewater treatment, J. Membr. Sci. 87 (1/2) (1994) 119–129.
[4] R.B. Bai, H.F. Leow, Microfiltration of activated sludge wastewater—the effect of system operation parameters, Sep. Purif. Technol. 29 (2) (2002) 189–198.
[5] R.B. Bai, H.F. Leow, Modeling and experimental study of microfiltration using a composite module, J. Membr. Sci. 204 (2002) 359–377.
[6] C. Chiemchaisri, Y.K. Wong, T. Urase, K. Yamamoto, Organic stabilization and nitrogen removal in membrane separation bioreactor for domestic wastewater treatment, Water Sci. Technol. 25 (10) (1992) 231–240.
[7] H. Nagaoka, S. Ueda, A. Miya, Influence of bacterial extracellular polymers on the membrane separation activated sludge process, Water Sci. Technol. 34 (9) (1996) 165–172.
[8] N. Park, B. Kwon, I. Kim, J. Cho, Biofouling potential of various NF membranes with respect to bacteria and their soluble microbial products (SMP): characterizations, flux decline, and transport parameters, J. Membr. Sci. 258 (2005) 43–54.
[9] M. Pribyl, F. Tucek, P.A. Wilderer, J. Wanner, Amount and nature of soluble refractory organics produced by activated sludge micro-organisms in sequencing batch and continuous flow reactors, Water Sci. Technol. 35 (1) (1997) 27–34.
[10] D.R. Noguera, N. Araki, B.E. Rittmann, Soluble microbial products (SMP) in anaerobic chemostats, Biotechnol. Bioeng. 44 (9) (1994) 1040–1047.
[11] I.S. Chang, C.H. Lee, Membrane filtration characteristics in membrane-coupled activated sludge system—the effect of physiological states of activated sludge on membrane fouling, Desalination 120 (1998) 221–233.
[12] J.S. Kim, C.H. Lee, I.S. Chang, Effect of pump shear on the performance of a crossflow membrane bioreactor, Water Res. 35 (9) (2001) 2137–2144.
[13] S. Comte, G. Guibaud, M. Baudu, Relations between extraction protocols for activated sludge extracellular polymeric substances (EPS) and EPS complexation properties. Part I. Comparison of the efficiency of eight EPS extraction methods, Enzyme Microb. Technol. 38 (2006) 237–245.
[14] S.R. Chae, S.T. Kang, Y. Watanabe, H.S. Shin, Development of an innovative vertical submerged membrane bioreactor (VSMBR) for simultaneous removal of organic matter and nutrients, Water Res., in press.
[15] S.R. Chae, H.S. Shin, Effect of condensate of food waste (CFW) on nutrient removal and behaviors of intercellular materials in a vertical submerged membrane bioreactor (VSMBR), Bioresource Technol., in press.
[16] G.J.F. Smolders, M.C.M. van Loosdrecht, J.J. Heijnen, Model of the anaerobic metabolism of the biological phosphorus removal process; stoichiometry and pH influence, Biotechnol. Bioeng. 43 (1994) 461–470.
[17] W.T. Lee, S.T. Kang, H.S. Shin, Sludge characteristics and their contribution to microfiltration in submerged membrane bioreactors, J. Membr. Sci. 216 (2003) 217–227.
[18] M. Taniguchi, J.E. Kilduff, G. Belfort, Modes of natural organic matter fouling during ultrafiltration, Environ. Sci. Technol. 37 (2003) 1676–1683.
[19] P. Gui, X. Huang, Y. Chen, Y. Qian, Effect of operational parameters on sludge accumulation on membrane surfaces in a submerged membrane bioreactor, Desalination 151 (2002) 185–194.
[20] B.E. Rittmann, W. Bae, E. Namkung, C.J. Lu, A critical evaluation of microbial product formation in biological processes, Water Sci. Technol. 19 (3/4) (1987) 517–528.
[21] R. Bura, M. Cheung, B. Liao, B.C. Finlayson, I.G. Lee, G.G. Droppo, S.N. Leppard, Composition of extracellular polymeric substances in the activated sludge floc matrix, Water Sci. Technol. 37 (4/5) (1998) 325–333.
[22] O.H. Lowry, N.H. Rosebrough, A.L. Farr, R.J. Randall, Protein measurement with the folin phenol reagent, J. Biol. Chem. 193 (1951) 265–275.
[23] B. Frølund, R. Palmgren, K. Keiding, P.H. Nielsen, Extraction of extracellular polymers from activated sludge using a cation exchange resin, Water Res. 30 (8) (1996) 1749–1758.
[24] M. Dubois, K.A. Gilles, K. Hamilton, P.A. Rebers, Colorimetric method for determination of sugars and related substances, Anal. Chem. 28 (1956) 350–357.
[25] P.C. Carman, Fluid flow through a granular bed, Transactions of the institution of chemical engineers 15 (1937) 150–167.
[26] F. Meng, H. Zhang, Y. Li, X. Zhang, F. Yang, Application of fractal permeation model to investigate membrane fouling in membrane bioreactor, J. Membr. Sci. 262 (2005) 107–116.
[27] P.R. Dugan, The function of microbial polysaccharides in bioflocculation and biosorption of mineral ions, in: Y.A. Attia (Ed.), Proceedings of the Flocculation in Biotechnology and Separation Systems, Elsevier Science Publishers B.V., The Netherlands, 1987, pp. 337–350.
[28] P.H. Hodgson, G.L. Leslie, R.P. Schneider, A.G. Fane, C.J.D. Fell, K.C. Marshall, Cake resistance and solute rejection in bacterial, microfiltration: the role of the extracellular matrix, J. Membr. Sci. 79 (1993) 35–53.