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Fouling reduction in MBR-RO processes: the effect of MBR F/M ratio

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ABSTRACT

This work investigated fouling propensities of microfiltration membranes and reverse osmosis (RO) membranes and analyzed the characteristics of deposited foulants on the membranes in two parallel Membrane bioreactor (MBR)-RO systems, with MBRs operated at different food to micro-organism (F/M) ratios. The results show that a high F/M ratio (0.50 g/g MLSS day) in the MBR caused greater membrane fouling rates of the MBR and RO membranes than a low F/M ratio (0.17 g/g MLSS day). In the MBRs, deposited microbial flocs were major foulants at low flux (10 L/m^2 h), whereas soluble substances in the cake foulants predominantly induced membrane fouling at high fluxes (20 and 30 L/m^2 h). In order to investigate the contributions of the protein, polysaccharides, and transparent exopolymer in the soluble substances to membrane fouling, bovine serum albumin, sodium alginate, and gum xanthan were used as model compounds, respectively, in experiments with the activated sludge samples from the MBR. The results imply that soluble polysaccharides (SP) and soluble transparent exopolymer particles (sTEP) were associated with fouling propagation. On the RO membranes, SP and sTEP were identified as major contributors to RO fouling rather than microbial cells and soluble protein. Our findings emphasize that the important role of the nature of soluble substances in membrane fouling and highlight that optimization of MBR operation is crucial to alleviate RO membrane fouling.

Keywords: Integrated membrane systems; Membrane fouling; Soluble polysaccharides; Transparent exopolymer particles; Extracellular polymeric substances; F/M ratio

1. Introduction

Membrane bioreactor (MBR)-reverse osmosis (RO) processes have been applied to reclaim municipal wastewater. An MBR process can remove above 95% organic carbon and completely remove suspended solids from wastewaters by biodegradation and membrane retention with less production of waste sludge.

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Subsequently, the RO membrane eliminates dissolved solids, organic compounds, nutrients, and pathogens in MBR effluent to produce high-quality reclaimed water [1–4].

However, MBR fouling and RO fouling, resulting in reduction in productivity and increases in energy costs, is still a major challenge in applications of MBR-RO systems. In MBRs, membrane-fouling propensity is associated with the characteristics of microbial flocs and soluble microbial products, which are determined by MBR operating conditions (e.g. sludge retention time (SRT), hydraulic retention time (HRT), substrate loading and composition, aeration, addition of flocculants, permeate flux, etc.). Meanwhile, the operating conditions of MBRs potentially influence the permeate quality. The colloidal, organic, and inorganic substances in MBR permeate promote organic fouling, biofilm growth, and scaling on RO membranes [5–7]. Accordingly, the RO membranes may display various performances under different operating conditions of MBRs.

Previous studies have pointed out that dissimilar food to micro-organisms (F/M) ratios could result in different microbial behaviors such as community shifts and activity variations, which potentially impact on biomass characteristics (e.g. concentration, particle size, viscosity, and floc structure) and extracellular polymeric substances (EPS) generations in MBRs [8]. Since microbial flocs and EPS are potential foulants, these variations could induce different fouling development trends in the MBRs.

Despite a number of studies have investigated the MBR and RO-fouling propensities of laboratory-scale or pilot-plant MBR-RO processes treating various wastewaters [1–3,5], detailed foulants characterization and dominant foulant identification (organic substances accumulation/biofilm growth) is rather limited towards effective control strategies. In addition, to date, there have been few studies on the effect of F/M ratio in the MBR on downstream RO membrane performance in MBR-RO processes.

This research aims to compare the fouling propensities of MBRs and RO membranes when the MBRs were operated at different ratios of food to micro-organisms (F/M) in the parallel MBR-RO process. In the each MBR, fluxes of 10, 20, and 30 $L/m^2 h$ (LMH) were employed for six modules, respectively, to explore predominant flux-dependent fouling contributors. In the RO systems, the contributions of soluble organic substances and viable cells in the RO biofilm layers to transmembrane pressure (TMP) increase were examined. The information on fouling behavior of the MBRs and RO membranes offers opportunities to reduce fouling in the MBR-RO processes by optimization of MBR operating conditions.

2. Materials and methods

2.1. MBR-RO description and operating conditions

A schematic of a laboratory-scale MBR-RO setup is shown in Fig. 1. Two MBRs were seeded with the activated sludge (~6 g/L) taken from a wastewater treatment and reclamation plant. The biomass concentrations in both MBRs were maintained at ~6 g/L by regulating the biomass wastage (i.e. adjusting SRT). A concentrated synthetic wastewater was fed into the two MBRs at an organic loading of 1.0 and 3.0 gCOD/L day, respectively, by regulating their individual feed pumps. At the same time, tap water was supplied into each MBR to maintain an effective reactor volume of 30 L, whose condition was controlled by a level sensor in the reactor. The composition of synthetic wastewater was CH_3COONa (32 g/L), NH_4Cl (4.8 g/L), $MgSO_4{\cdot}7H_2O$ (0.4 g/L), K_2HPO_4 (3.5 g/L), $CaCl_2{\cdot}2H_2O$ (0.55 g/L), and $FeSO_4{\cdot}7H_2O$ (0.02 g/L). Each MBR had six submerged flat-sheet membrane modules (polyvinylidene fluoride (PVDF), hydrophilic, 0.08 μm nominal pore size, 0.0288 m^2 of effective surface area for each membrane module, Toray Industries, Inc., Japan). An instantaneous permeate flux of 10, 20, or 30 LMH was obtained by regulating the flow rate of individual suction pumps (on 9 min/off 1 min, controlled by a timer). The TMP of each membrane module was monitored by a pressure transducer, which was connected to a personal computer equipped with a data logging system (Msystem, Japan). Hydrochloric acid was automatically added into the reactor when the pH was higher than 7.1, whose condition was controlled by a pH sensor in the reactor.

Two parallel RO cells with commercial brackish water RO membranes (UTC-70, 32 mm × 7 mm, 0.0186 m^2 of effective surface area, Toray Industries, Inc., Japan) and feed channel spacers were used. For each RO unit, the MBR permeate was collected and stored in a feed tank (10 L) with a stirrer (IKA, Germany) at a temperature of 25 ± 1 ℃ (controlled by a cooling water system). The MBR permeate was delivered from the feed tank using a high-pressure pump (Winston Engineering Corporation, Singapore) to the RO cell. The feed pressure was controlled by a back-pressure regulator (Swagelok, USA) and the feed and permeate pressures were monitored by pressure transducers (Ashcroft, USA). The cross-flow rate of feed (20 L/hr, equivalent to 0.1 m/s) was regulated by a flow control valve (Swagelok, USA) and recorded by a flow meter (Brooks Instrument, USA). The permeate flow rate was monitored and controlled by a mass flow controller (Brooks Instrument, USA) to maintain the permeate flux of 20 LMH automatically. The conductivities of feed and permeate were measured by

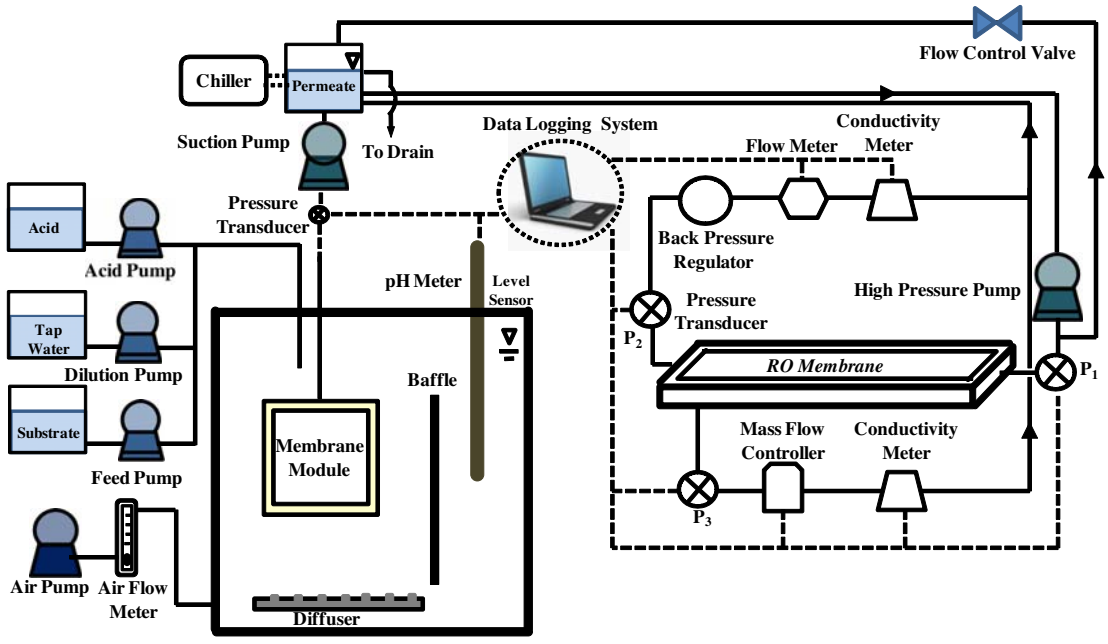

Fig. 1. Schematic diagram of the MBR-RO system.

conductivity meters (Thermo Scientific, USA). Due to the limited productivity of MBR permeate, the RO concentrate and RO permeate were recycled back to the feed tank and overflow of this mixture was conducted. The pressure transducers, mass flow controllers, and conductivity meters were connected to a computer equipped with data logging system (LabVIEW, National Instruments, USA) [9,10].

2.2. Analytical methods

Mixed liquor suspended solids (MLSS) in the MBR were measured by Standard Methods [11]. Microbial floc size was examined with a laser particle sizer (Malvern, UK). The pH was measured by a pH meter (Horiba, Japan).

2.3. MBR permeate quality evaluation

The dissolved organic carbon of the MBR permeate sample was monitored using a total organic carbon (TOC) analyzer (Shimadzu, Japan) after the sample was pretreated with a filter (PTFE, hydrophilic, 0.45 μm, Advantech, Japan). The EPS amount in the MBR permeate sample was examined by analyzing polysaccharides and protein contents, according to the methods described by Dubois et al. [12] and Bradford [13], respectively. Transparent exopolymer (TEP) content was measured based on the method first introduced by Passow and Alldredge [14]. In summary, the sample was filtered through a polycarbonate filter (Millipore, USA) with a pore size of 0.1 μm (therefore, we are reporting TEP > 0.1 μm). The accumulated TEP on the filter were subsequently stained with 3 mL of 0.02% aqueous solution of alcian blue in 0.06% acetic acid. Then, the filter was washed with distilled water to remove the excess dye. Six milliliters of 80% H_2SO_4 solution was used to remove the complex of TEP and alcian blue from the polycarbonate membrane and allowed to dissolve it for 2 h. The absorption of the solution was measured at a wavelength at 787 nm using a spectrometer (Hach, USA), and the concentration of TEP was calculated based on a calibration with gum xanthan as a standard. The chemicals used in the EPS and TEP experiments were from Sigma-Aldrich (USA).

2.4. Sampling of foulants

The virgin microfiltration membrane was put into a tank filled with distilled water and its TMP was monitored (defined as TMP_m) before being used for the MBR. When the TMP reached a certain level (defined as TMP_{total}), the membrane module was taken from the MBR and then was replaced by a new membrane. The foulants were gently removed from the membrane surface using a cotton stick and suspended with 15 mL of distilled water (herein defined as MBR foulant solution). The physical-cleaned membrane was put into a tank filled with distilled water and the TMP was measured (defined as TMP_{i+m}). The cake layer fouling

(defined as TMP_c, i.e. reversible fouling) was determined by the difference of TMP_{total} and TMP_{i+m}, and the irreversible fouling (defined as TMP_i) was calculated by subtracting TMP_m from TMP_{i+m}. The averaged membrane-fouling rate (kPa/day) was calculated as $(TMP_{total} - TMP_m)$/filtration time.

The virgin RO membrane was installed in the RO system. The averaged RO-fouling rate (kPa/day) was determined as $[(TMP_t - \Delta\Pi_t) - (TMP_0 - \Delta\Pi_0)]/t$, where TMP presents the applied pressure across the membrane, $\Delta\Pi$ osmotic pressure difference between the RO feed and permeate, and t filtration time [10]. At different fouling stages, the membrane was removed from the RO system and a new RO membrane replaced it. The RO foulants were obtained from the fouled RO membrane using a cotton stick. The foulants were then suspended with 15 mL of distilled water (herein defined as RO foulant solution) for further analysis.

2.5. Characterization of foulants

2.5.1. Dry weight

The total weight of MBR and RO foulants was measured as proposed by Standard Methods [11].

2.5.2. Adenosine triphosphate (ATP)

Viable cells in the foulants derived from the RO membranes were evaluated by measuring the intracellular ATP concentrations [15,16] following the manufacturer's manual. In summary, after mixing the sample with eliminating extracellular ATP reagent (Kikkoman, Japan) for 30 min, the sample was added into a test tube (Kikkoman, Japan) containing 100 μL of ATP releasing agent solution (Kikkoman, Japan). After 20 s, 100 μL of luciferin-luciferase (Kikkoman, Japan) was added into the tube and the amount of light (Relative light unit (RLU)) produced was measured using a luminometer (Lumitester C-110, Kikkoman, Japan). The intracellular ATP concentration was calculated using the conversion factor for the linear relationship between RLU values and concentrations of ATP standard solutions. The total ATP concentration was measured following the same procedure without adding the eliminating reagent. The extracellular ATP concentration (indicating dead cells) was determined by the difference between total ATP concentration and intracellular ATP concentration.

2.5.3. EPS

The pellet and supernatant were separated from the foulant sample by centrifuging at 1,780 × g for 10 min at 4°C. The bound EPS was extracted from the pellet following a "formaldehyde-NaOH" method described previously [17]. The polysaccharides and protein contents in the bound EPS (from the pellet) and soluble EPS (from the supernatant) were examined according to the methods described by Dubois et al. [12] and Bradford [13] respectively.

2.5.4. Soluble TEP

TEP in the soluble RO foulants were measured based on the Passow and Alldredge method [14].

2.6. Bench-scale filtration test

A bench-scale filtration assay (Fig. 2) was set up to investigate the contributions of soluble protein, polysaccharides, and TEP to membrane fouling in experiments with activated sludge mixed liquor from the MBR. Bovine serum albumin, sodium alginate, and gum xanthan (Aldrich-Sigma, USA) were used as model compounds representing protein, polysaccharides, and TEP, respectively. The characteristics of the model compounds are shown in Table 1 and Fig. 3. The model foulant solution was prepared by dissolving a certain amount of the model compound into distilled water. A membrane (PVDF, hydrophilic, 0.08 μm, 0.001 m^2 of surface area, Toray Industries, Inc., Japan) was put in a 100-mL filtration cell. The operating pressure was maintained at 5, 15, and 25 kPa provided by an air compressor. The magnetic stirrer bar (a rotation speed of ~350 rpm) was installed in the filtration cell and located at 0.5 mm above the membrane surface. The permeate flux was determined by weighting the permeate on an electronic balance (A&D company, Japan), which was connected to a personal computer equipped with a data logging system (A&D company, Japan).

The membrane filtration resistance was calculated according to the Darcy's equation, $R = \Delta P/(\mu J)$, where ΔP is the driving pressure (25 kPa), μ is the water viscosity, and J is the filtration flux (i.e. the permeate

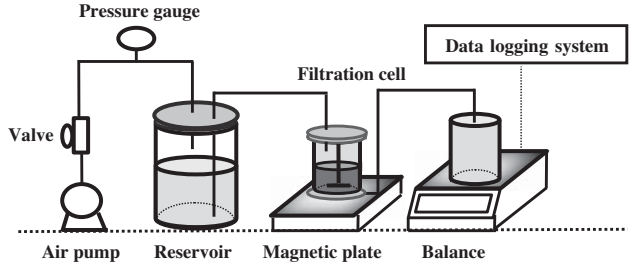

Fig. 2. Schematic diagram of bench filtration setup.

Table 1
Properties of model foulants and their mixtures with activated sludge

		Bovine serum albumin, fraction V	Sodium alginate from brown algae	Gum xanthan from *Xanthomonas campestris*
Model foulant	Manufacturer	Sigma-Aldrich	Sigma-Aldrich	Sigma-Aldrich
	Average molecular weight (Dalton)[a]	67,000	–	–
	Carbon content (g TOC/g)[b]	0.44 ($R^2 = 0.996$)	0.29 ($R^2 = 0.999$)	0.28 ($R^2 = 0.989$)
	pH @ 25°C (750 mg/L in H_2O)[c]	6.2 ± 0.1	6.7 ± 0.1	5.6 ± 0.1
Mixed with activated sludge[c,d]	pH @ 25°C	7.8 ± 0.1	8.0 ± 0.1	7.7 ± 0.1
	Particle size d_{50} (μm)	178 ± 8	177 ± 5	171 ± 10
	Zeta potential (mV)	−25 ± 4	−23 ± 1	−28 ± 3

[a]Data from manufacturer.
[b]$n = 6$.
[c]$n = 3$.
[d]The activated sludge was taken from the high F/M-MBR. The pH of activated sludge was 7.8 ± 0.1, the particle size of activated sludge was 182 ± 6 μm, and zeta potential of activated sludge was −22 ± 2 mV. The mixture was consisted of 10 mL of 750 mL model compound and 10 mL of activated sludge.

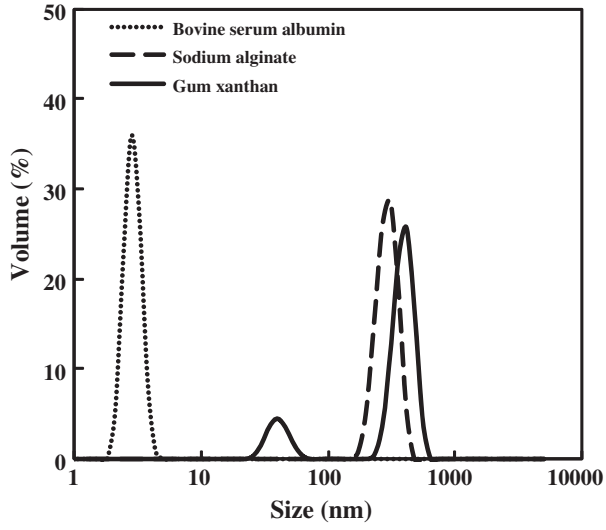

Fig. 3. Size distribution profiles of model foulants.

production rate specific to membrane filtration area). Initially, distilled water was filtered to monitor the clean membrane resistance (R_m). The sample (10 mL of activated sludge from the high F/M-MBR with 10 mL of distilled water or with 10 mL of model foulant solution) was then placed in the filtration cell and filtered for 30 min. The sample volume was maintained by replenishing with distilled water from a reservoir. The normalized resistance was achieved by calculating $R_{(as+model\ foulant)}/R_{as}$, in which $R_{(as+model\ foulant)}$ represents the averaged resistance of activated sludge with model foulant solution at filtration time of 28 ~ 30 min and R_{as} means the averaged resistance of activated sludge with distilled water at filtration time of 28 ~ 30 min.

3. Results and discussion

3.1. Effect of F/M ratio on MBR fouling

When the MBR approached the steady state, the high F/M-MBR achieved an averaged SRT of ~7-day and the low F/M-MBR had an averaged SRT of ~45-day by maintaining the MLSS at about 6 g/L in the reactors and controlling discharge sludge amounts.

In this study, when the TMP reached ~25 kPa, the fouled membrane module was taken from the reactor and replaced by a new membrane module. The averaged membrane-fouling rates at fluxes of 10, 20, and 30 LMH in both reactors after 2 × SRT are given in Table 2. It is apparent that the low F/M-MBR had relatively slower membrane-fouling rate (5~20-fold less) compared with the high F/M-MBR at the three fluxes employed. The results also indicated that cake layer fouling was predominant fouling (80–90% of the total fouling) in both MBRs. Furthermore, the cake layer foulants were removed from the membranes and the accumulation rates of bound EPS and soluble EPS in the foulants were examined.

Table 2
The effect of F/M ratios in the MBRs on fouling rates of MBRs and RO membranes[a]

		Low F/M ratio (0.17 g/g day)	High F/M ratio (0.50 g/g day)
MBR fouling rate (kPa/day)	Flux of 10 LMH	0.04	0.84
	Flux of 20 LMH	0.72	3.36
	Flux of 30 LMH	1.2	13.7
RO fouling rate (kPa/day)		22	115

[a]The datum is the averaged value which is calculated based on data from the multi operations ($n = 2 \sim 5$).

Fig. 4(A) shows that at a flux of 10 LMH, greater accumulation rates of bound EPS derived from microbial flocs (0.29 ± 0.10 and 1.29 ± 0.38 mg/g foulants day for the low F/M-MBR and high F/M-MBR, respectively) were found in the foulants than those of soluble EPS (0.09 ± 0.01 and 0.74 ± 0.01 mg/g foulants day for the low F/M-MBR and high F/M-MBR, respectively), which reveals that the microbial flocs/developed biofilms were predominant contributors to cake formation in both MBRs. At a flux of 20 LMH, slightly more soluble EPS (5.6 ± 2.9 and 5.1 ± 2.0 mg/g foulants day for the low F/M-MBR and high F/M-MBR respectively) were deposited on the membranes compared with the bound EPS (1.5 ± 1.2 and 3.7 ± 1.9 mg/g foulants day for the low F/M-MBR and high F/M-MBR, respectively). Further increasing the flux to 30 LMH, considerably higher accumulation rates of soluble EPS (about 4 times) than those of bound EPS in both MBRs. This implies that the role of soluble EPS in forming cake layers and inducing membrane fouling became more significant with elevating filtration flux (Fig. 4(B)).

In the MBR, mixed liquor includes microbial flocs, colloids including planktonic bacteria, and macrosolutes (EPS, etc.). The aeration provided to the reactor generates cross-flow, which induces back transport of foulants and determines the critical flux. The larger flocs experience shear-induced diffusion, which increases with particle size, whereas the submicron colloids and macrosolutes experience Brownian back diffusion, which decreases with their size; there tends to be a minimum critical flux around 0.1 micron and critical flux would tend to be higher for the supra-micron flocs [18]. The consequence of this is that at typical MBR conditions of bubbling, filtration (9 min) and relaxation (1 min) in this study, the microbial flocs do not tend to form a cake, but the colloids and EPS macrosolutes are preferentially converted to form a foulant layer. Over time the foulant layer could include bacterial microcolonies mainly derived from recruited planktonic bacteria.

Recently, the influence of transparent exopolymer particles (TEP) on membrane fouling in MBRs has also received increasing attention [19]. To further compare the contributions of soluble EPS (mainly polysaccharides and protein) and soluble TEP to membrane fouling in the MBR, a bench-scale constant pressure stirred dead-end filtration study using model foulant com-

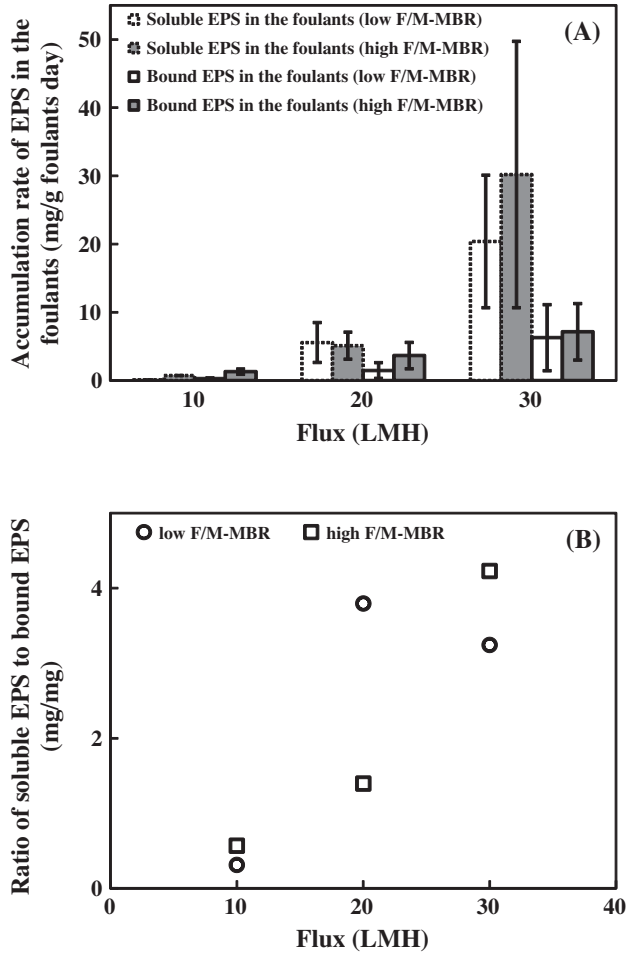

Fig. 4. Accumulation rates of EPS (A) and ratios of soluble EPS to bound EPS (B) in the foulants at various fluxes.

pounds (bovine serum albumin, sodium alginate, and gum xanthan, 10 mL) with activated sludge mixed liquor (10 mL) from the MBR was performed. It is noted that additions of model foulant compounds at experimental concentrations would not significantly affect the pH value, particle size, and surface charge of the activated sludge mixed liquor (Table 1).

The bench-scale filtration tests at TMP of 5, 15, and 25 kPa were performed in order to explain the contributions of the model foulants to membrane fouling at various fouling stages. Fig. 5 demonstrates that the normalized resistances of the activated sludge-model foulant (BSA, alginate, and gum xanthan) mixtures were almost independent of the applied pressures.

Moreover, addition of BSA (up to 750 mg/L) into the activated sludge samples from both MBRs could not exacerbate membrane fouling (Fig. 5(A)). A similar phenomenon was observed by Wang and Waite that protein could not affect membrane-fouling rate due to their nonstructural role in the gel layers [20]. Addition of alginate at a concentration of 75 mg/L into the activated sludge samples from both MBRs did not significantly accelerate the development of membrane fouling. However, when the alginate concentration in the mixture was raised to 750 mg/L, slight resistance increases (~1.2-fold and ~1.4-fold for the activated sludge samples from the low F/M-MBR and high F/M-MBR, respectively) were observed. Additions of the gum xanthan (75 mg/L) into activated sludge samples from both MBRs resulted in increases of filtration resistance about 1.2-fold. Interestingly, the low F/M-MBR and high F/M-MBR responded differently to gum xanthan at a high concentration (750 mg/L). Significant increases in normalized resistances (~2.7-fold) were found when the gum xanthan (750 mg/L) were mixed with the activated sludge from the high F/M-MBR compared to that from the low F/M-MBR (~1.5-fold). Compared with alginate, gum xanthan induced faster normalized resistance increase, especially at a high concentration. This hints that the soluble TEP could play an important role in inducing membrane fouling in the MBR.

3.2. Effect of F/M ratio on RO fouling

After about 150-day operation of MBRs, the amounts of DOC, protein, polysaccharides, and TEP in the permeates of both MBRs were compared (Table 3). It is evident that the high F/M-MBR pro-

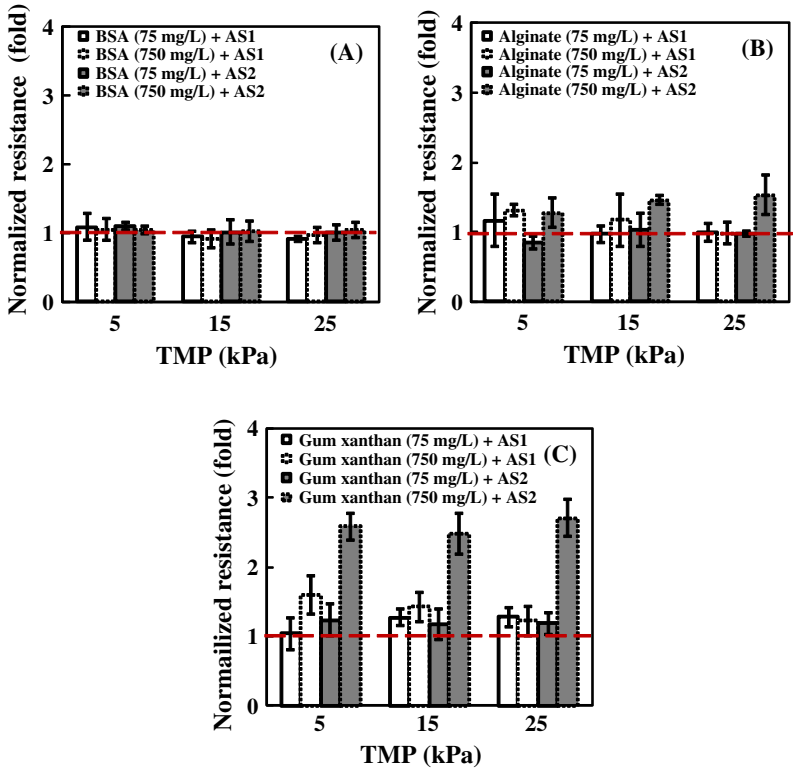

Fig. 5. Contributions of model foulants to resistance increases in activated sludge from different F/M-MBRs at various TMP levels. AS1: the activated sludge from the low F/M-MBR; AS2: the activated sludge from the high F/M-MBR.

duced permeates with higher DOC, polysaccharides, and TEP contents than the low F/M-MBR (Table 3). The MBR permeates were then fed to the parallel RO systems and the results show that the high F/M-MBR permeate induced higher fouling rate (~5-fold) compared with the low F/M-MBR (Table 2). This was attributed to higher organic substances (indicated as polysaccharides, TEP, and DOC) in the high F/M-MBR permeate compared with that in the low F/M-MBR permeate (Table 3), which contributed to increase the accumulated organic layers (e.g. nonbiodegradable organic substances) and facilitate the biofilm propagation (e.g. biodegradable organic substances) on the RO membranes.

On the RO membranes, the directly deposited substances from the MBR permeate and developed biofilms utilizing the substances from the MBR permeate contributed to RO fouling. To further identify the major contributors to aggravate membrane performances, soluble organic substances (polysaccharides, protein, and TEP) and biofilm growth (viable cells) in the RO foulants were examined. Their accumulation rates on the membranes were calculated and are shown in Fig. 6. It was found that the soluble TEP and soluble polysaccharides (SP) (derived from MBR permeate, excretion substances of viable biofilm cells, and lysis substances of dead biofilm cells) accumulated on the RO membrane were strongly correlated with the RO fouling rate (Fig. 6(B) and (D)). It is worth noting that there is still lack of sufficient evidence to explain the relative contributions of viable cells and soluble protein to RO fouling (Fig. 6(A) and (C)). To further identify the dominant contributors at various fouling stages, time-dependent organic substance deposition and bacterial growth on RO membranes needs to be further examined.

Table 3
Effect of F/M ratio on MBR permeate quality

	Low F/M-MBR	High F/M-MBR
DOC (mg/L)	4.5 ± 2.5	11.7 ± 6.3
Protein (mg/L)	8.4 ± 6.3	5.5 ± 3.7
Polysaccharides (mg/L)	2.9 ± 1.8	14.9 ± 8.0
TEP (mg gum xanthan/L)	1.7 ± 1.3	5.1 ± 4.0

3.3. Implication of this study

3.3.1. Fouling control strategies at different fluxes in MBRs

In this study, the results indicated that the ratios of microbial flocs (indicated by the bound EPS) in the cake layer foulants dropped with elevating

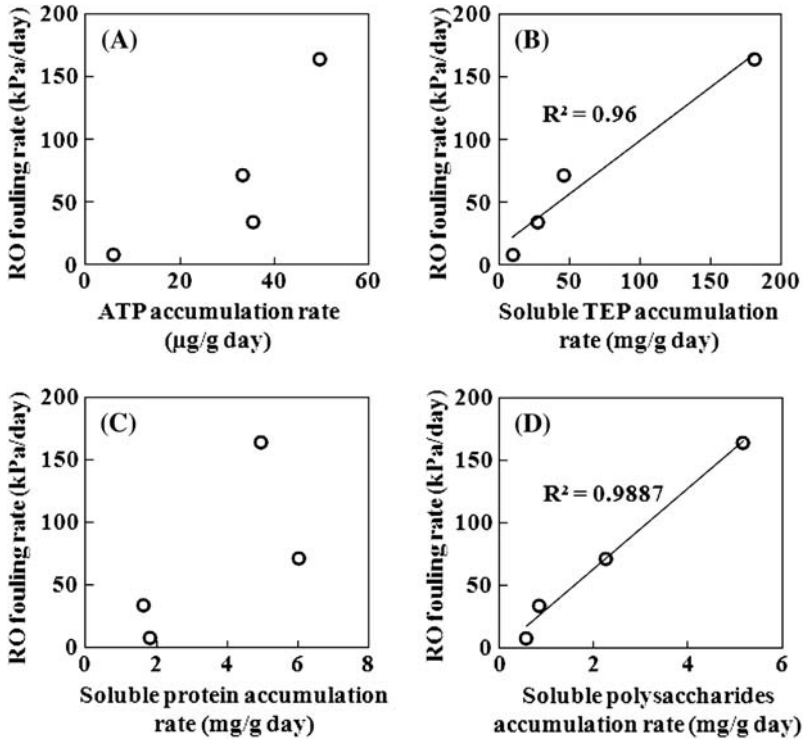

Fig. 6. Relationship between the accumulation rates of viable cells and soluble substances in the RO foulants and RO fouling rate.

filtration flux, but the ratios of soluble substances (indicated as the soluble EPS) in the cake layers became more dominant with an increase of flux. This implies that various membrane-fouling control strategies may be considered at different filtration fluxes (i.e. different dominant foulants). Physical cleaning (e.g. backwashing) or using chemicals (e.g. energy uncoupling reagents) could greatly limit fouling at a low flux by inhibiting microbial floc depositions and biofilm growth [21]. While, at a high flux, the addition of coagulants or flocculants into the MBRs to adsorb or coprecipitate with soluble substances could be effective strategies to control membrane fouling caused by soluble substances (such as EPS or TEP) [22].

3.3.2. Optimization of MBR operating conditions to reduce membrane fouling in MBR-RO systems

In this study, we observed that the low F/M ratio in the MBR not only alleviated membrane fouling of MBRs, but also produced the permeate with less organic substances, which induced less RO fouling. Interestingly, dramatically increasing the model foulants contents in the activated sludge from the low F/M-MBR did not significantly cause more serious membrane fouling (Fig. 5). This may be attributed to the structural nature of microbial flocs in the low F/M MBR, which could facilitate the soluble substances entrapped into their matrix rather than their contact with the membrane surface. This finding implies that MBR operating conditions determine RO-fouling development and optimization of MBR operation is a crucial approach to alleviate RO fouling.

Because less membrane fouling happened in the low F/M-MBR, the total cost for membrane cleaning (including the chemical cost, the cleaning manpower cost, as well as the cost equivalent to decreasing operation ratio of equipment and increasing frequency of membrane replacement, etc.) would be greatly lower than that in the high F/M-MBR. Moreover, the slower TMP increase in the low F/M-MBR could lead to relatively less energy consumption of the suction pumps compared with the high F/M-MBR. However, it is noted that at the same HRT and biomass concentration, the treating capability of the low F/M (0.17 g/g day, equivalent to a loading of 1.0 g COD/L day) –MBR-RO was 2-times lower than that of the high F/M (0.5 g/g day, equivalent to a loading of 3.0 g COD/L day)-MBR-RO. Thus, the operating cost and energy requirements need to be evaluated when the operating conditions of MBRs are optimized to reduce membrane fouling.

3.3.3. TEP, an alternative parameter to relate with fouling development

TEP was initially investigated as a major agent in the aggregation of particles in the fields of oceanography and limnology. With the development of TEP measurement protocols, TEP can be distinguished from the EPS and other organic substances based on its reaction with Alcian Blue. Therefore, recently, the influence of TEP on membrane fouling of MBRs and RO membranes has received increasing attention [23,24].

TEP consists of gel-like acidic fractions of polysaccharides, proteins, and nucleic acid. The sticky characteristics of TEP could cause them to adhere to membrane surfaces, and also mobilize bacteria onto membranes. Moreover, the bacteria embedded in the matrix could utilize the organic substances as nutrients to promote their propagation and to excrete microbial products on the membranes [19,25,23,26]. A number of studies have reported the roles of TEP in inducing membrane fouling, however, its significance is still being debated. For example, whether the developed TEP measurement protocols are suitable to be used in water treatment monitoring is still not clearly known. The studies on the relationship between TEP and EPS are still in the early stage. It is believed that future study on the behaviors of TEP in influencing membrane fouling will allow us better understanding membrane fouling mechanisms in MBRs and RO membranes.

4. Conclusions

The F/M ratio of the MBR had influences on fouling tendencies of the MBRs and RO membranes. Higher fouling propensities of microfiltration and RO membranes were observed in the high F/M-MBR-RO system. In the MBRs, soluble substances (protein, polysaccharides, and TEP) were identified as major foulants and their contributions to membrane fouling typically followed the order: TEP > polysaccharides > protein. In the RO systems, the accumulations of SP and TEP on the RO membranes were strongly linked to fouling development. This study emphasizes the importance on optimizing MBR operating conditions (i.e. at a low F/M ratio) to minimize RO fouling by producing less organic substances (e.g. polysaccharides and TEP) in the permeate.

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