



Impact of fouling index behavior in RO fed at different MBR operating conditions in MBR-RO

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ABSTRACT

The purpose of this study is to compare fouling index of reverse osmosis fed in MBR-RO systems at different operating conditions in MBRs. Lab. scale reactor of MBRs was operated at different flux and temperature and permeate was measured SDI₁₅ to predict RO fouling rate. The results suggest that formed cake layer in membrane surface, higher flux and lower temperature in MBRs, led to lower RO fouling rates and SDI₁₅ value was decreased, because of cake layer attached greater amounts of organic substances such as polysaccharide and protein.

Keywords: Wastewater reuse; MBR-RO process; MBR operating condition; Cake layer; Fouling index; Silt density index

1. Introduction

As water shortages are increasing, the need for sustainable water treatment and there use of water is essential. The dual-membrane process, micro-filtration (MF) or ultra-filtration (UF) and reverse osmosis (RO), sharply increases as a technology for the reclamation of municipal wastewater because of its efficiency and easy and economical operation [1,2]. Especially, the MBRs can remove above 95% organic carbon and completely remove suspended solids from biological treatment process instead of sedimentation. For the above reasons, there is a growing interest in the MBR-RO process because of the pretreatment of RO (Reverse osmosis) process was not required such as MF (Micro filtration) or UF (Ultra filtration).

However, the membrane processes are generally limited by membrane fouling, which reduces productivity and increases maintenance operation costs. In case of MBR-RO process, RO membrane fouling was the main reason [6]. As RO feed water, MBR effluent was composed of various materials such as the colloidal, organic, extracellular polymer particle and inorganic substances, was promoted

organic fouling and scaling of RO membranes. In addition, the MBRs permeate quality could be influence by the operating conditions (e.g., hydraulic retention time (HRT), sludge retention time (SRT), F/M ratio (i.e., food to microorganisms ratio) and composition, flux, membrane pore size, etc. that determine biomass characteristics (e.g., concentration, viscosity, microbial community, EPS production) and/or membrane fouling conditions. For example, according to Kent et al. found that proteins were the predominant RO foulants in the initial fouling stage of the RO membrane, but polysaccharide deposition on the membrane surface became dominant after operating the RO membrane for a few weeks in a MBR-RO system [9]. Additionally the MBR permeate of high and low F/M ratio in MBRs was also related with the fouling behaviors of the RO membranes [10]. Therefore, the operating condition of MBR effected on the RO membrane fouling in MBR-RO. However, to date, there have been few studies on the effect of MBR operating conditions on downstream RO membrane performance. The relationship between MBR fouling tendency by operating condition and RO feed characteristics is also not well understood. This research aims to predict the fouling pro-

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propensities of RO membrane fed with MBRs permeate through SDI_{15} value, which obtained from lab scale reactor and SDI_{15} measurement device at different operating conditions (temperature and flux) in MBRs. The relationship between MBRs permeate quality and fouling propensities of MBRs with operating conditions was investigated. The contribution of MBR fouling propensities with operating conditions (temperature and flux) to SDI_{15} values were examined. The information on SDI_{15} value of the RO membranes feed offers opportunities to reduce RO fouling in the MBR–RO processes by operating conditions in MBRs.

2. Materials and methods

2.1. Experimental set-up and membrane

This study was performed with lab scale submerged membrane bioreactor of 100 L volume and PES flat sheet modules (Microdyn-nadir, Germany) as shown in respective Table 1 and Fig. 1. This was controlled using a water bath and automatically operated at constant flow rate by PLC program. This study purpose was the effect of RO fouling tendency with MBRs operating conditions in the MBR–RO systems and the experimental conditions were summarized in Table 2.

Table 1
Characteristics of MBR membranes applied in lab scale reactor

Parameter	Method
Manufacturer	Microdyn-nadir Inc. (Germany)
Module type	Flat sheet
Material	Polyethersulfone (PES)
Pore size (μm)	0.04
MWCO (kDa)	150
Membrane surface (m^2)	0.34

2.2. Analysis of membrane fouling model

From filtration theory based on the Darcy's equations:

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

where J is the membrane permeate flux ($\text{m}^3/\text{m}^2\text{h}$), TMP is the trans-membrane pressure (Pa), μ is the viscosity of permeate ($\text{Pa}\cdot\text{s}$), R_t is the total resistance ($1/\text{m}$). The following individual resistances were selected to describe the resistance-in-series [Eq. (2)]. Considering fouling buildup the mechanism, R_a can be calculated the membrane resistance after physically backwashing with de-ionized water (DIW) from the R_t' according to Eq. (3).

$$R_t = R_m + R_c + R_p + R_a \quad (2)$$

$$R_t' = R_t - R_p = R_m + R_a \quad (3)$$

R_m (virgin membrane resistance) determined from measuring the water flux of DIW using virgin membrane. R_t was calculated by the recorded data at the end of filtration operation. A temperature correction to 20°C was used to account for the dependence of permeate viscosity, according to the following equation [11].

Table 2
Operating conditions of lab scale MBRs

Parameter	Mode 1	Mode 2
Temp ($^\circ\text{C}$)	10, 15, 20, 25 & 30	20
Flux ($\text{L}/\text{m}^2\cdot\text{h}$)	30	10, 20, 30 & 40
SADam	0.55	
MLSS conc. (mg/L)	17,540 (16,800–18,420)	

^aSADm represents specific aeration demand with respect to the membrane area.

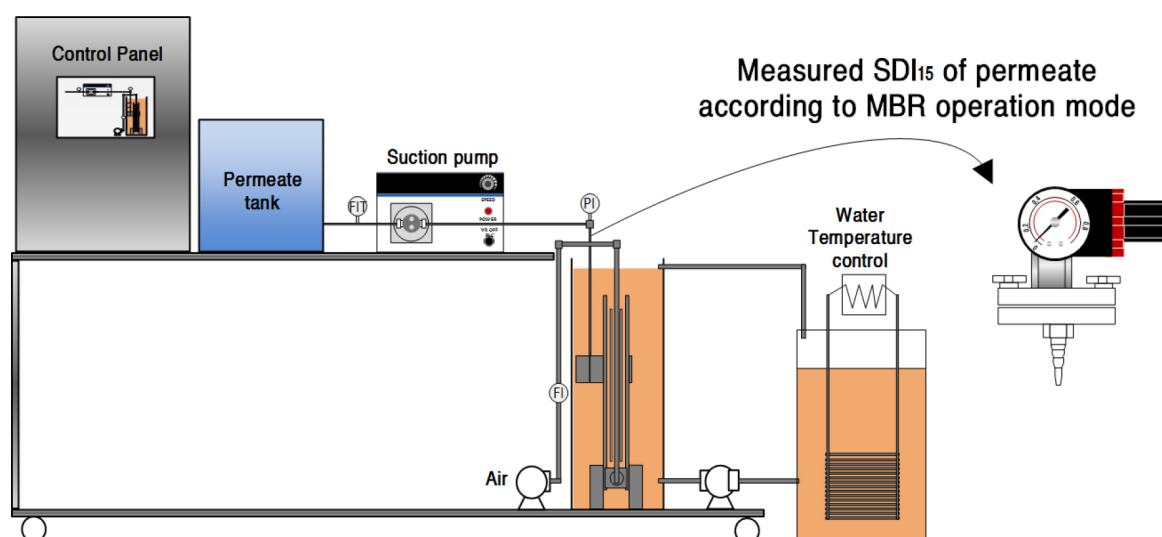


Fig. 1. Schematic diagram of lab. scale reactor and SDI_{15} apparatus.

$$J_T = J_{20} \times 1.024^{(T-20)} \quad (4)$$

2.3. Extraction of EPS

The extraction of EPS was performed by the thermal extraction method in this study [14]. Protein analysis was measured by Bradford Assay method and polysaccharide was measured by phenol sulfate method. All the above analyses were conducted in duplicate, and their average values were reported.

2.4. FEEM (Fluorescence excitation-emission matrix) analysis

A Shimadzu spectrofluorophotometer (RF-6000) was used to measure FEEM in this experiment. The peak intensity values were determined by setting the slit interval between excitation and emission at 5 nm, wavelength range between excitation and emission from 200 to 400 nm and from 250 to 600 nm. Based on the values obtained by measuring the peak intensity of the wavelength combination between excitation and emission in which the maximum fluorescence intensity was obtained from each specimen, the analysis was conducted using the method developed by Chen et al. According to Chen et al., it was possible to divide the FEEM results into five regions for evaluation. Regions I, II, III, IV, and V could be represented by aromatic protein, aromatic protein, fulvic acid-like substances, soluble microbial by-product-like substances, and humic acid-like substances, respectively [15].

2.5. SDI definition and method for RO fouling index

The fouling index has been functioned as one of the most useful tools to predict fouling potential of membrane process. A good fouling index can indicate how rapidly given feed water will foul the RO membrane system based on the fouling mechanisms. Association for Testing and Materials (ASTM D 4189–95) [16], providing standard method of its measurement as well as the manner of application. Measurement of the SDI followed the standard test method using a 0.45 mm MF filter (Millipore, Billerica, CA). The SDI measurement equipment consisted of a membrane holder, a high-pressure pump controlled by a pressure control system, a feed water reservoir and a data

acquisition system. The applied hydraulic pressure was maintained at 2.07 ± 0.07 bar and the feed water temperature was maintained at $20 \pm 1^\circ\text{C}$. The SDI can be calculated by Eq. (5) :

$$SDI = \left(1 - \frac{t_i}{t_f}\right) \times \frac{100}{t_T} \quad (5)$$

where T is total test in min (usually 15 min), t_i is time (sec) to collect initial 500 mL of sample and t_f is time (in sec) to collect final 500 mL of sample.

3. Results and discussion

3.1. Variation in characteristics of MBRs fouling with operating temperature

In order to derive the characteristics of the membrane fouling in MBRs with temperature in the MBR-RO system, proceeded to the Mode 1 of Table 2.

The results of this study showed that the mean values of the filtration resistance over time were as shown in Fig. 3 as $-1.895\text{E}-12$, $1.677\text{E}-12$, $-1.363\text{E}-12$, $-1.158\text{E}-12$ and $-9.709\text{E}-13$ 1/m, and the fraction of fouling resistance ratio (R_c/R_p) was 0.012, 0.037, 0.055, 0.106 and 0.174, respectively. It was found to be main cause of cake layer formation on the membrane surface. In addition, concentration of $\text{EPS}_{\text{bound}}$ was measured to investigate the main cause of cake layer at different temperature in MBRs, and results presented Fig. 4.

The concentrations of $\text{EPS}_{\text{bound}}$, protein and polysaccharide, were increased from 177.2 mg/L to 869.1 mg/L and 64.8 mg/L to 89.8 mg/L respectively as temperature decreased 30 to 10°C . Especially, protein increase was higher than polysaccharide and it was considered that it had a direct effect on formation of cake layer due to concentration increase of $\text{EPS}_{\text{bound}}$ from Biomass released as lower temperature, which was in correspondence with the results of Wang et al. [17]. For example, Jang et al. [18] reported that the increase in filtration resistance was directly related to SMPs or $\text{EPS}_{\text{bound}}$ concentration. In addition, Chang and Hernandez Rojas [19,20] reported protein as the main factor of membrane fouling.

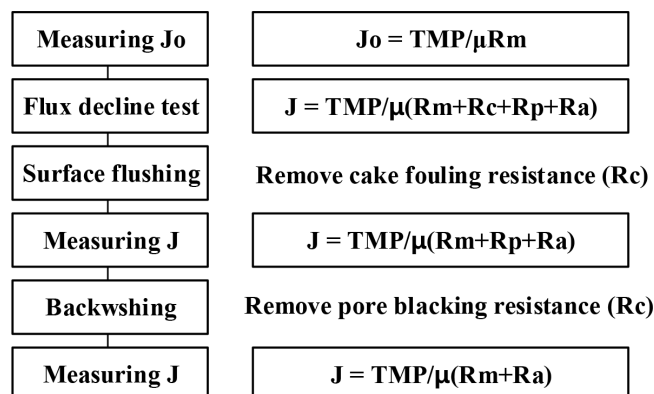


Fig. 2. Analytical method of fouling model.

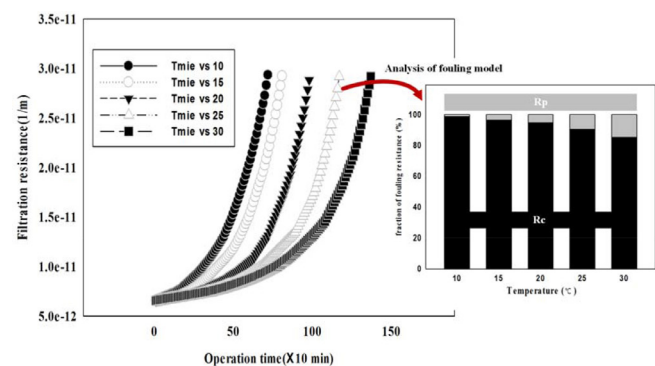


Fig. 3. Comparison results of filtration resistance and fraction ratio of fouling resistance with operating temperature.

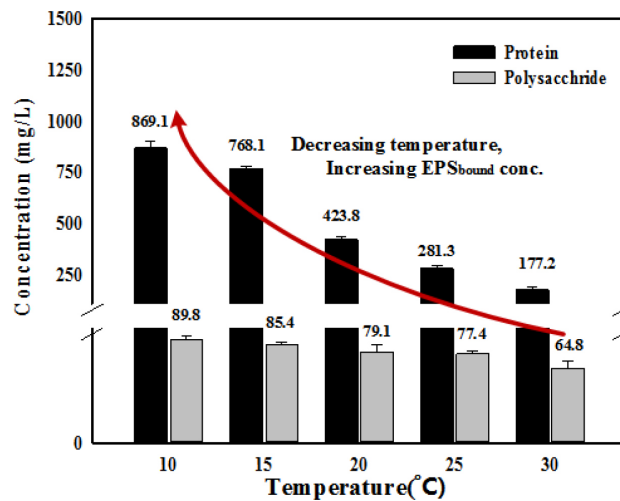


Fig. 4. Comparison results of protein and polysaccharide with operating temperature.

3.2. Variation in characteristics of RO feed at with operating temperature in MBRs

To investigate the effect of RO feed at operating temperature in MBRs, protein and polysaccharide were measured in MBRs permeate. As presented in Fig. 5, the concentration of EPS_{bound} , protein and polysaccharide, dropped as temperature increases. Whereas the concentration of them in MBRs permeate increased. In these results, it indicated the RO feed caused by cake layer formation was identified as the dominant contributor to reduce fouling factor by adsorbing on membrane in MBRs. Based on the above experimental results, the samples of MBRs permeate at temperature were analyzed by FEEM and were illustrated in Fig. 6.

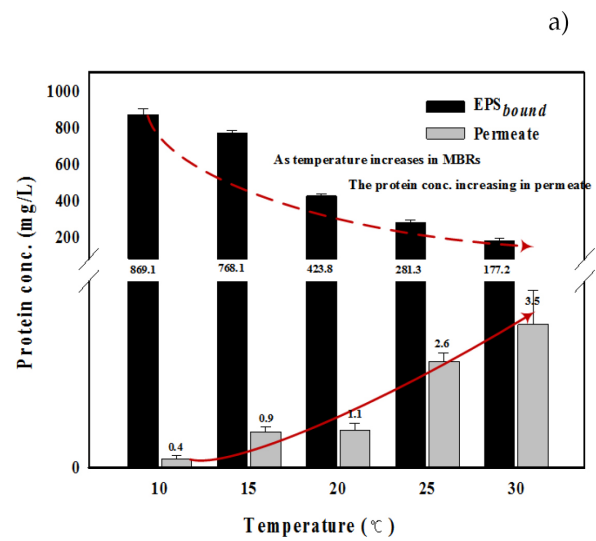
As the water temperature increased, the main peak points of MBRs permeate were observed in regions II, IV and V, and especially the variation of humic acid and aromatic protein were remarkable. These results show that the removal efficiency of RO fouling factor, substrate like humic acid and protein, due to the adsorption on the cake layer is decreased as the temperature is increased and the MBRs process is closely related to the cake layer formation.

3.3. Variation in characteristics of MBR permeate with operating flux

The membrane fouling characteristics of MBRs with operating flux proceeded to Mode 2 of Table 2, and the results showed that the mean values of the filtration resistance over time were as presented in Fig. 7 as $2.28E-12$, $1.58E-12$, $1.36E-12$ and $1.08E-12$ 1/m respectively.

To identify the membrane fouling, the fraction of fouling resistance (R_c/R_p) was 0.114, 0.085, 0.037 and 0.018 respectively. These results have been leading main cause of membrane fouling by forming cake layer on membrane similar to results of Mode 1.

According to Miller et al. [21], as the flux in the MBRs increases, the amount of fouling was attached to the membrane surface increases more than can be removed by physical methods such as shear force. In this study, formation of



b)

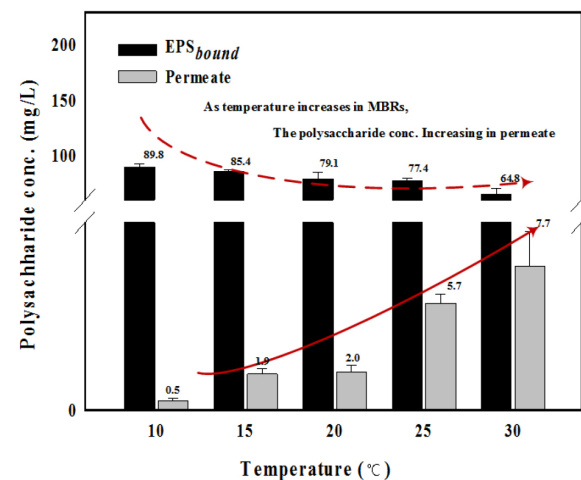


Fig. 5. Comparison concentration of EPS_{bound} and permeate, protein (a) and polysaccharide (b), in MBRs with operating temperature.

cake layer on membrane surface was the main cause as the flux increased in MBRs.

3.4. Variation in characteristics of MBR permeate with flux

To investigate the effect of RO influence at operation flux in MBRs, concentration of protein and polysaccharide, were examined. As presented in Fig. 8, SMPs concentration of MBRs permeate was dropped as flux increased. In these results, it was indicated to the variation of fouling factors in RO feed caused by cake layer formation in the MBRs. Which was identified as the dominant contributor to reduce fouling factor by adsorbed on membrane in MBRs. Especially, the removal efficiency of protein was high because of the high molecular weight.

As presented in Fig. 9, the results of the FEEM analysis showed a tendency to decrease in the in regions II, IV and

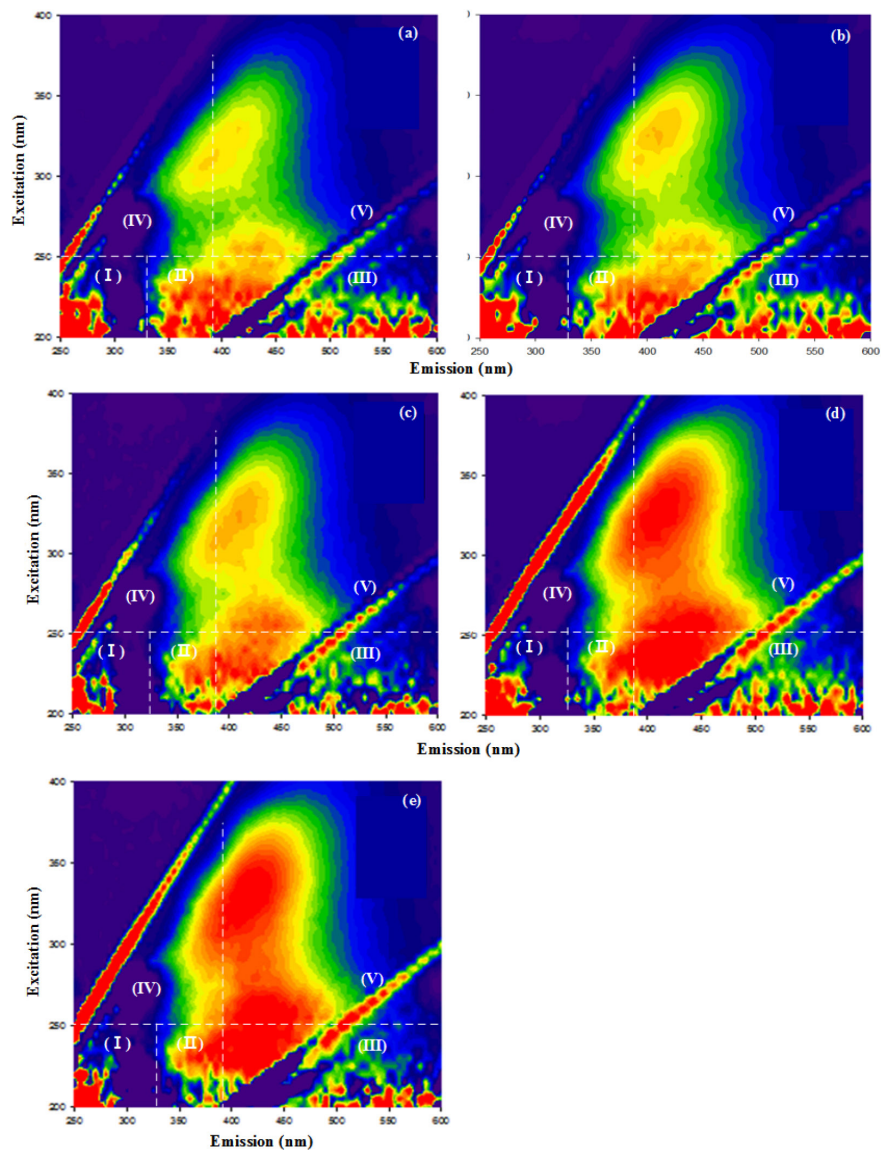


Fig. 6. FEEM fluorescence spectra of soluble microbial products on variation of temperature with operating conditions; (a) 10°C, (b) 15°C, (c) 20°C, (d) 25°C, (e) 30°C.

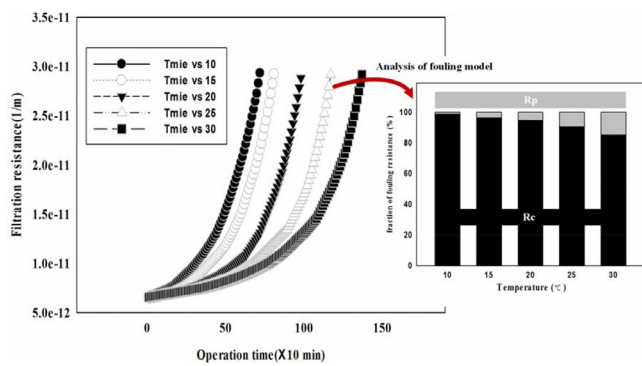


Fig. 7. Comparison results of filtration resistance and fraction of fouling resistance with operating temperature.

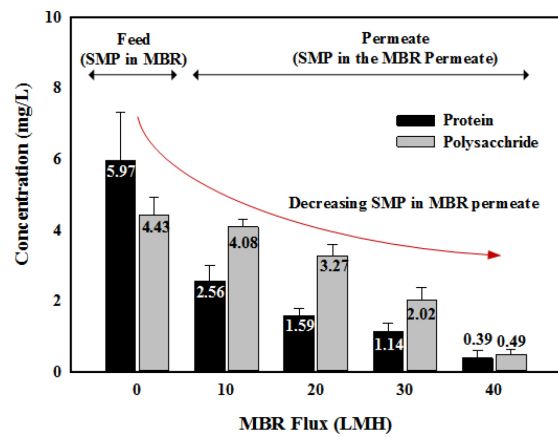


Fig. 8. Comparison concentration of EPS_{bound} and permeate, protein (a) and polysaccharide (b), in MBRs with operating flux.

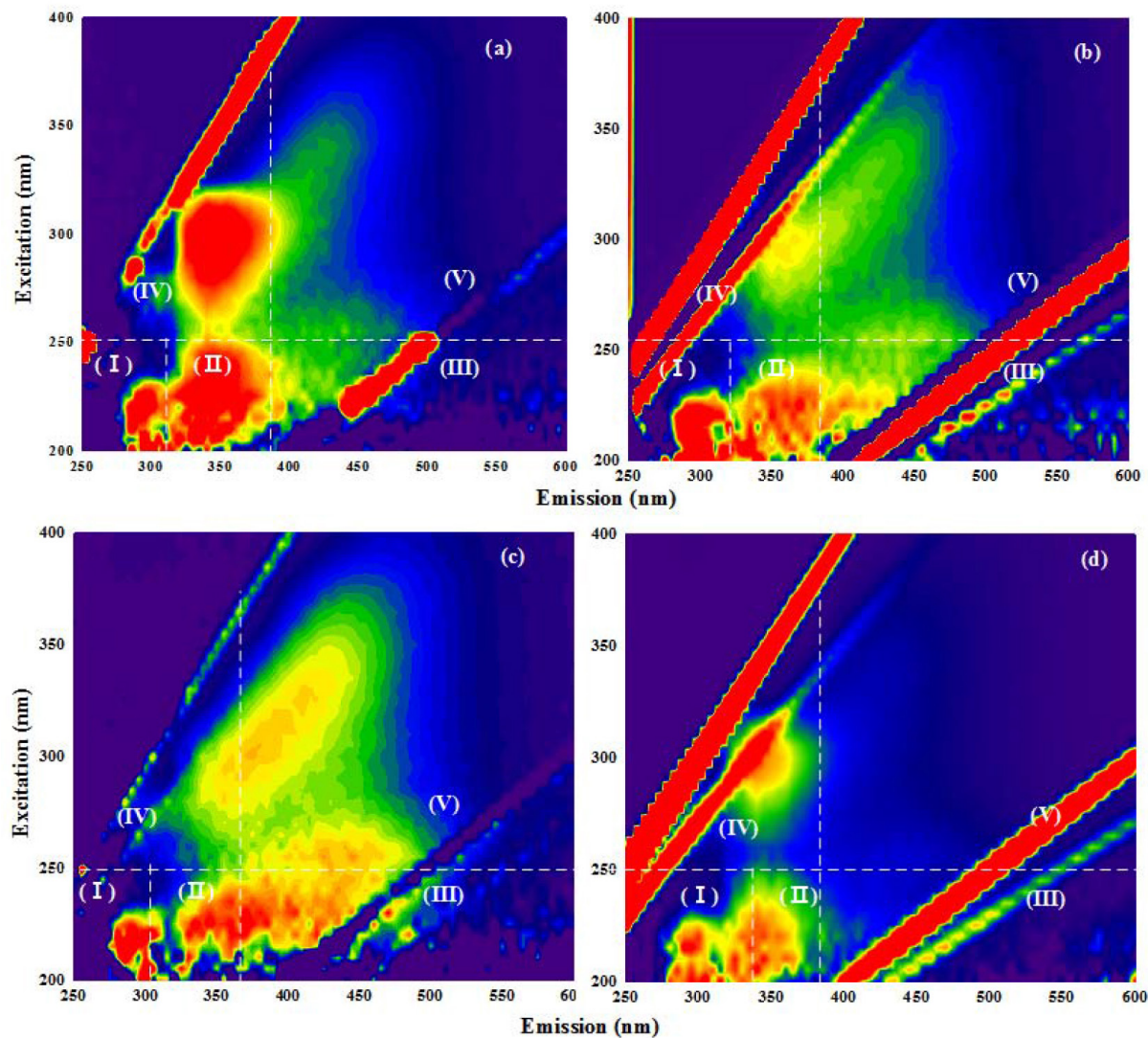


Fig. 9. FEEM fluorescence spectra of MBRs permeate on variation of flux with operating conditions; (a) 10 LMH, (b) 20 LMH, (c) 30 LMH, (d) 40 LMH.

V, especially it was observed that operating condition of increasing flux in MBRs had a lower fluorescence intensity in regions II (aromatic protein II) and IV (soluble microbial by-product-like). These results that the accumulation of protein and polysaccharide tended to increase as the cake layer thickened, which was correspondence with results of Dimitios C. et al. [22].

3.5. Evaluation of RO fouling index in the MBR-RO with operating conditions in the MBRs

In order to predict RO membrane fouling in the MBR-RO system, the SDI_{15} of the MBRs permeate with operating conditions, temperature and flux in the MBRs, was measured. As presented in Fig. 10a, SDI_{15} values of MBRs permeate were increased from 2.14 to 4.33 as temperature risen and Fig. 10(b) showed that it was decreased from 4.20 to 2.48 as flux increased in the MBRs. It could

also be closely associated with the formation of cake layer on membrane surface in MBRs and these results showed that SDI_{15} value of RO fed was decreased as the cake layer thickened. This explanation is supported by F.C. Kent et al., [10] who observed that proteins made up a higher proportion of fouling layers initially after which the polysaccharide deposition densities increased dramatically. The organic matter accumulation increased by developing the cake layer on the membrane surface in MBRs, which was correspondence with results of Dimitios C. et al. [22].

4. Conclusions

This study investigated the fouling prediction of RO membrane on the MBR operating conditions (i.e., flux and temperature) through SDI_{15} value measurement. The results showed that the MBR process operating at higher flux and

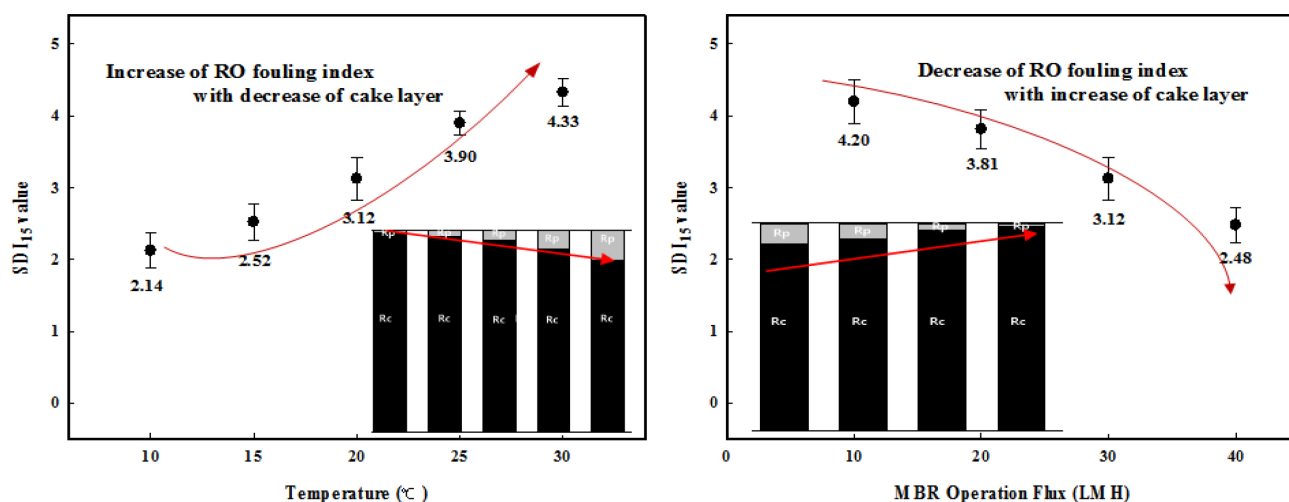


Fig. 10. Comparison SDI₁₅ values of MBRs permeate with operating conditions.

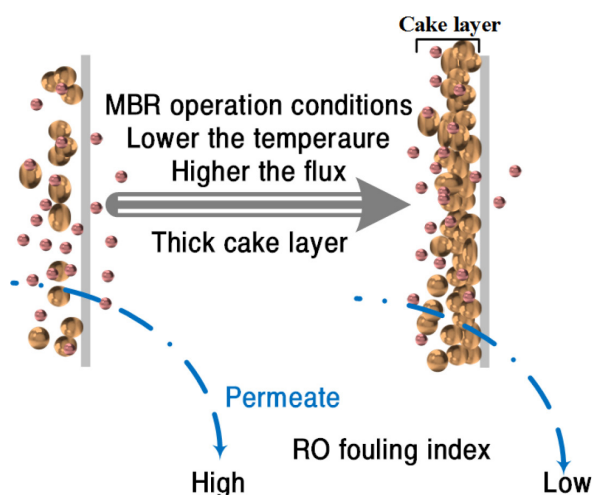


Fig. 11. Schematic representation of variation on RO fouling index according to MBRs operating conditions in MBR-RO system.

lower temperature had lower SDI₁₅ values of feed to RO membrane. These results were demonstrated through the MBR fouling propensities and the most closely related with the occurred cake layer at operating conditions of MBRs. The thicker the cake layer on membrane surface in MBRs was adsorbed more fouling factor, polysaccharide and protein, of RO membrane feed, therefore the SDI₁₅ value seen to be dropped. This study implies that operating conditions of MBRs is a crucial strategy to reduce RO membrane fouling in the MBR-RO system.

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