# Evaluation of CEB strategies on irreversible fouling control at low temperature in MBRs

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### ABSTRACT

The purpose of this study was to investigate the effect of microbial characteristics on the membrane fouling with temperature and to derive effective membrane fouling control by CEB at low temperature under TMP point. The measurement of membrane fouling with temperature was performed considering the viscosity of water. Our results are as follows. As the temperature decreased, the increase of EPS concentration from microorganisms was the main cause. Especially, polysaccharide concentration was closely related to irreversible fouling than protein concentration in MBRs. In addition, we selected pressure of cleaning point by deriving TMP jump point to control of irreversible fouling at low temperature, CEB was carried out with changing condition of cleaning point and chemical dosage. As a result, it was confirmed that the CEB efficiency dropped sharply at above TMP jump point. These results suggest that different cleaning protocol of membrane fouling with temperature, by using chemically enhanced backwashing, not only in appropriate cleaning methods of physical and chemical, but also in those times.

Keywords : Membrane bioreactors; Low-temperature; Irreversible fouling control; Chemical enhanced backwash; TMP Jump

#### 1. Introduction

Membrane bio reactors (MBRs) have gained increasing popularity in municipal, domestic, and industrial wastewater treatment [1–3]. Compared to conventional activated sludge processes, using MBRs has the advantage of having a smaller environmental footprint, excellent effluent quality, and less sludge production [4]. One major disadvantage of MBR is the fouling that occurs during operation. The fouling raises trans-membrane pressure (TMP), which results in additional energy consumption. Many papers have been devoted to exploring the properties of foul ants, factors influencing fouling and fouling mechanisms, and fouling mitigating strategies [5,6]. Thus far, membrane cleaning in MBRs has not been adequately addressed. This is due to the

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following reasons: (1) membrane fouling, being a very complicated issue, is not yet fully understood; (2) fouling and cleaning issues are specifically related to membrane properties, feed-biomass characteristics, and operating conditions; (3) cleaning protocols in full-scale MBRs are typically recommended from membrane manufacturers and/or MBR suppliers, and some cleaners are proprietary [8]. The membrane fouling has been studied with regard to the effect of operation parameters, such as sludge retention time (SRT), temperature [9], pH and DO concentration [10].

In Korea, there are four seasons and the influent temperature of sewage treatment plant is reported to be 8 to 30°C. Especially in the winter season, membrane fouling is rapidly progressed, and frequent chemical cleaning is required, which adds to the difficulty in the operation of the process. Therefore, there is a demand for a protocol for controlling the membrane fouling characteristics with

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temperature [11]. According to previous studies, Research has found that the concentration of extra celluar polymeric substances (EPS) and soluble microbial products (SMPs), a well-known substance that causes membrane fouling [12], was increased as temperature decreased. These materials are composed of a variety of organic substances that was released from microorganisms as the result of their metabolic activity and biomass decay. However, the cleaning protocol of membrane fouling at low temperature is limited. Therefore, a cleaning protocol suitable for the field situation is required.

The purpose of this study was to investigate the effect of microbial characteristics on the membrane fouling with temperature and to derive effective membrane fouling control by CEB at low temperature. Membrane fouling characteristics were investigated in lab. scale MBR system and the cleaning efficiency was measured according to operating pressure and chemical concentration to control membrane fouling at low temperature condition.

#### 2. Materials and methods

#### 2.1. MBRs description and operating conditions

A Schematic of a lab scale MBRs setup is presented Fig. 1. The lab scale reactor was constructed to maintain the temperature during the experiment using water bath, and the process was controlled at a constant flow rate through a PLC control program. The MBR had a submerged flat-sheet membrane modules (Polyethersulfone, (PES), 0.04 µm nominal pore size, 0.34 m<sup>2</sup> of effective surface area, Micro dyn, Inc., Germany). A permeate flux was maintained for membrane module by regulating the flow rate of suction pumps (on 9 min/off 1.5 min, controlled by a timer). The TMP of membrane module was monitored by a pressure transducer, which was connected to a laptop equipped with data logging system. The MBR operating conditions are summarized in Table 1.

#### 2.2. Determination of filtration resistance

Different types of membrane filtration resistances were applied to evaluate the degree of membrane fouling. The degree was calculated using the following resistance-in-series model and Darcy's equations:

$$R_t = R_m + R_f + R_c \tag{1}$$

$$Rt = \frac{TMP}{(\eta_T J)} \tag{2}$$

where *J* is the membrane permeate flux (m/h), *TMP* is the trans-membrane pressure (Pa),  $\eta_T$  is the viscosity of permeate (Pa·s),  $R_i$  is the total resistance (1/m),  $R_m$  is the intrinsic membrane resistance (1/m),  $R_c$  is the cake layer resistance,

Table 1 Operating conditions of the lab scale MBRs

Parameter	Value	
HRT (h)	8 h	
Flux (LMH)	40	
Filtration mode	9 min on/1 min off	
Designed F/M ratio (g	0.57	
CODcr/g MLSS d)	(0.50~0.61)	
SRT (d)	0.04	
Temperature	10, 15, 20, 25 and 30 (°C)	
Backwashing rate	1Q	
Cleaning point	0.10, 0.15, 0.20 and 0.25 k $gf/cm^2$	
CEBa		
Chemical concentration	200, 400, 600, 800 and 1,000	
	mg/L as NaOCl	
	0.05 N as NaOH	



Fig. 1. Schematic diagram of lab scale MBRs.

and  $R_f$  is the fouling resistance (1/m) caused by irreversible adsorption and pore blockage.

 $R_m$  was determined from measuring the water flux of Milli-Q water using an original 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 [13].

$$\eta_T = \eta_{20} \cdot e^{(T-20)} \tag{3}$$

where the temperature (T) is in Celsius.

For the calculation of  $R_m + R_p$  we rinsed the fouled membranes with tap water to drain off the cake layer. We then filtrated Milli-Q water through the membrane to determine  $R_m + R_f$  by using Eq. (2).  $R_c$  was evaluated by subtracting  $R_m + R_f$  from the  $R_{t'}$  according to Eq. (1).

#### 2.3. FEEM (fluorescence excitation-emission matrix) analysis

We used a Shimadzu spectrofluorophoto meter (RF-6000) to measure FEEM. The peak intensity values were determined by setting the slit interval between excitation and emission at 5 nm, and 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, we conducted our analysis 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. These regions were as follows: region I - aromatic protein; region II - aromatic protein II; region III - fulvic acid-like substances; region IV - soluble microbial by-product-like substances; and region V – humic acid-like substances [14].

### 2.4. Soluble microbial products (SMP) analysis and bound extracellular polymeric substances (EPS)

We collected 20 ml sludge samples 10 times from both MBRs. We stored the samples in a 4°C refrigerator overnight before measuring. The SMP and EPS<sub>bound</sub> were characterized by their relative content of protein and carbohydrate. We centrifuged the samples at 4,000 rpm for 5 min. The supernatant were then filtrated by a filter with a pore size of 0.22 µm. We used permeates for SMP analysis, while we used the settled solids for further preparation of bound EPS samples. A heating method described by Le-Clech, et al. [5] was used as the extraction method for bound EPS. The settled solid was mixed with 20 ml Milli-Q water and heated by a water bath at 80°C for 10 min. The samples were then centrifuged at 7,000 rpm for 10 min. The supernatant were filtrated by a filter with a pore size of 0.22 µm. Permeates were then ready for the bound EPS analysis. We measured the proteins and carbohydrates samples of the SMP and the bound EPS by a spectrophotometer (Bio-Rad Benchmark Plus, USA) at 490 nm and 595 nm, respectively. We based these methods on the Bio-Rad protein assay for the proteins (Bio-Rad Laboratories Inc., USA) and the phenol-sulfuric acid method for the carbohydrates [15,16].

### 3. Results and discussion

### 3.1. Characteristics of filtration resistance with temperature condition of MBR process

To proceed this experiment, the flux of the lab scale MBR system was determined considering the viscosity of water with temperature. In addition, the activated sludge used in the experiment was sampled from the sludge of the S university sewage treatment plant which was acclimated to influent of water temperature. Experiments were also conducted at 40 LMH using a preliminary critical flux test. The experimental temperature condition was set at 10, 15, 20, 25, and 30 degrees in consideration of the influent water temperature of the sewage treatment plant in Korea. As shown in Fig. 2, the filtration resistance in consideration of the viscosity of water increased as temperature decreased. It indicated the rate of increasing filtration resistance caused by variation of microbial characteristics with temperature, was also identified as the dominant contributor to membrane fouling, which was in accordance with the finding of Zhun Ma (2013) [17]. Therefore, we analyzed EPS and FEEM in order to investigate the effects of microbial influences on membrane contamination with changes in water temperature.

## 3.2. Effect of activated sludge behavior and membrane fouling propensity with temperature condition.

EPS<sub>bound</sub> and EPS<sub>soluble</sub> are composed of a complex matrix of proteins, polysaccharides, lipids and nucleic acids. These materials are also well known to have a direct impact on membrane fouling in MBRs. In addition, the sampling points were selected 2 points, feed (sludge in MBRs) and permeate, in order to investigate the effect of membrane fouling from sludge characteristics with temperature. As presented in Fig. 3, feed concentrations of both polysaccharide and protein increased as temperature dropped. Especially, protein was higher than polysaccharide, which was in correspondence with the results of van den Brink et al. [19]. However, permeate concentration of the polysaccha-



Fig. 2. Result of filtration resistance with temperature of operating condition.

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Fig. 3. Comparison results of protein and polysaccharide, feed and permeate, with operating temperature.

ride concentration was higher than that of protein, and this result suggests that the protein is easier to remove from the membrane due to its higher molecular weight than the polysaccharide.

Based on the above experiments, FEEM (Fluorescence excitation-Emission Matrix) analysis was conducted to examine whether EPS materials were adsorbed on the membrane. As shown in the Fig. 4, influent and effluent water were compared, and the adsorption levels of foulants were observed by the chromatic difference at the peak. As a result of analysis, it was considered that main cause of membrane fouling propensity were the change of microorganism characteristics with temperature.

# 3.2.1. Evaluation of irreversible membrane fouling by temperature conditions

To investigate the effect of temperature variation on membrane fouling, the filtration resistance before and after backwashing was measured, and its results and was shown in Table 2. As the water temperature decreased from 30°C to  $10^\circ \text{C},$  the rate of irreversible fouling increased from 5.12%to 11.02%. The rate of irreversible fouling increase was similar to the rate of EPS increase from microbial with temperature variation. As shown in Fig. 5, the highest rate of EPS release from microbial was observed when the protein was from 25°C to 20°C and the poly saccharide was from 15°C to 10°C. Also, it was confirmed that poly saccharide concentration in EPS material is closely related to irreversible fouling in MBRs than protein concentration. The poly saccharide has a relatively low molecular weight than the protein and is thought to have directly affected irreversible fouling, fast pore narrowing or pore blocking explained the high fouling irreversibility, which was in correspondence with the results of van den Brink et al. [19]. Therefore, we tried to derive the cleaning efficiency according to the CEB condition after estimating the TMP point as a method to efficiently control irreversible fouling which increases rapidly at low temperature.

## 3.3. Derivation of CEB conditions for control of irreversible fouling at low temperature.

### 3.3.1. Selection of pressure range to efficient chemical enhanced backwashing

In order to control of irreversible fouling at low water temperature conditions, we selected the proper pressure of cleaning point considering TMP jump. We decided four operating pressures, -0.10, -0.15, -0.20, -0.25 and -0.30 kgf/ cm<sup>2</sup>, by deriving the cleaning point as shown in the Fig. 7.

## 3.3.2. Evaluation of irreversible fouling control at low temperature by CEB

On the basis of previous experiments, CEB condition selected four points: 0.10, -0.15, -0.20, -0.25 and -0.30 gf/ cm<sup>2</sup>. Moreover, we selected chemical concentrations (NaOCl 200 mg/L, 400 mg/L, 600 mg/L, 800 mg/L, 1,000 mg/L, and 0.05 N NaOH) in consideration of adverse effect of microbial activity during backwashing (1 Q).

As presented in Fig. 7, cleaning efficiency of CEB increased as pressure lower and chemical concentration higher. In particular, it was confirmed that the rate of recovery rapidly decreased at above the TMP points (-0.30 kgf/ cm<sup>2</sup>). These results indicate that irreversible fouling can be controlled through the CEB under the TMP jump section, Which was in correspondence with the result of Keneko (2013) who mentioned chemical cleaning must be carried out with chemical reagents after a given period of processing time, when the transmembrane pressure (TMP) exceeds a given value, because some foulants cannot be removed.

In addition, as shown in Fig. 9, the recovery rate of the filtration resistance according to the NaOCl dosage and



(e) FEEM analysis (at 30  $^{\circ}$ C) EPS<sub>soluble</sub> (left), effluent water (right)

Fig. 4. FEEM fluorescence spectra of soluble microbial products on variation of temperature, (a)  $10^{\circ}$ C, (b)  $15^{\circ}$ C, (c)  $20^{\circ}$ C, (d)  $25^{\circ}$ C, (e)  $30^{\circ}$ C.

Table 2
Recovery rate of filtration resistance after backwashing

Backwash recovery rate					
Mode		Initial filtration resistance (1/m) after backwashing 30 s (1/m)	Filtration resistance	Filtration resistance Rate of increase (%)	
40 LMH	30°C	7.96E–11	8.39E-11	5.12	
	25°C	7.70E–11	8.19E-11	5.98	
	20°C	7.44E–11	7.99E–11	6.88	
	15°C	6.84E–11	7.53E–11	9.16	
	10°C	6.94E-11	7.80E–11	11.02	



Fig. 5. Comparison of EPS concentration and increasing rate of irreversible fouling with temperature variation.



Fig. 6. Results of TMP jump range selection via lab scale reactor.

the pressure of cleaning point was derived as an exponential function form (Recovery rate =  $113.27 \times \exp(-5 \times ((x - 0.152)/0.1365)^2 + ((y - 1239.74)/1322.49)^2))$ ). From this equation, it is thought that the recovery rate can be predicted according to the NaOCl dosage and pressure of cleaning point when operating MBR in the field.



Fig. 7. Compare of recovery ratio of filtration resistance that the effect of chemical dosage with cleaning point pressure.





Fig. 8. Correlation analysis of recovery ratio of filtration resistance with pressure of cleaning point and NaOCl dosage.

### 4. Conclusions

The purpose of this study was to investigate the effect of microbial characteristics on the membrane fouling with temperature and to derive effective membrane fouling control by CEB at low temperature under TMP point. The measurement of membrane fouling with temperature was performed considering the viscosity of water. Our results are as follows. As the temperature decreased, the increase of EPS concentration from microorganisms was the main cause of membrane fouling in MBRs. Especially, polysaccharide concentration was closely related to irreversible fouling than protein concentration. In addition, we selected CEB condition of pressure of cleaning point, considering TMP point, to control of irreversible fouling at low temperature, CEB was carried out with changing condition of cleaning point and chemical dosage. As a result, it was confirmed that the CEB efficiency dropped sharply at above TMP jump point. Consequently, CEB condition under TMP jump point should be taken into account when operating MBRs at low temperature.

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