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Biofilm formation and its effect on biofouling in RO membrane processes for wastewater reuse

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ABSTRACT

To reuse wastewater effluents, several advanced technologies including reverse osmosis (RO) membrane processes are being considered. However, biofouling caused by biofilm formation on the surface of RO membranes impedes their application for wastewater reclamation. Since the adhesion of bacterial cells to mambrane surfaces is the first step of biofouling, in this study, the bacterial adhesion tendency on two different surfaces, glass and RO membrane, was compared to investigate biofouling potential using two feed water sources with different organic content and ionic strength. The experimental results of biofilm formation potential showed that bacterial adhesion was substantially high when wastewater effluent was continuously supplied to the RO membrane surface due to physicochemical interactions including surface roughness and feed water properties. The membrane biofouling was determined by measuring of flux decline patterns and by analyzing foulants on the membrane surfaces. The rapid flux decline in the RO membrane exposed to the wastewater effluent was likely to be due to the biofilm formation in terms of extracellular polymeric substances (EPSs). Chlorination, the common biofouling control method, was conducted to reduce bacterial adhesion potential and remove the EPSs from the RO membrane surface. Neveretheless, the permeate flux was not improved and the EPS concentration was not decreased with increasing chlorine doses when the wastewater effluent was supplied.

Keywords: Biofouling; Reverse osmosis membrane; Wastewater effluent reuse; Extracellular polymeric substances

1. Introduction

Interests in reclamation and reuse of wastewater effluents have increased over the past two decades due to shortages of fresh water and increases of water demand in many regions of the world. Effluents from municipal wastewater treatment plants have become a reliable water source because of the large volume and the continuous supply. However, the municipal wastewater effluents contain high concentrations of organic matters and microorganisms; therefore, advanced technologies are required to meet the strict regulatory guidelines. Membrane processes, especially reverse osmosis (RO) membrane processes, have expanded their applications for reclamation and reuse of municipal wastewater effluents [1,2].

Membrane fouling caused by deposition of inorganic particulates, adsorption of organic matters, and microbial

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adhesion and growth, however, causes operational problems. As a result, extensive pretreatments are required to control the membrane fouling. Biofouling due to biofilm formation on the membrane surface is known to be difficult to control by pretreatment methods, since some microorganisms can survive and rapidly re-grow after the pretreatment. Biofouling is thus recognized as the most serious problem in RO membrane plants [3–6].

The first step in membrane biofouling is the deposition of bacterial cells to membrane surfaces [4]. The deposition of microorganisms on solid surfaces can be considered as a two-step process: (1) the particles are transported close to the adhesive surface, and (2) adhesion takes places under the control of physicochemical interactions and shear forces [7]. It is known that greater hydrophobicity of cells and substrata results in greater attractive forces and higher levels of adhesion, whereas more negative surface charges of cells and lower ionic strength result in greater repulsive electrostatic interactions and lower levels of adhesion [7]. Kang et. al investigated deposition rates of microorganisms on membrane surfaces and reported that the deposition were predominantly controlled by electrostatic double layer repulsion and permeation drag [8]. Consequently, control of bacterial adhesion and biofilm formation are needed to successfully apply the RO membrane processes for wastewater reuse.

In addition, extracellular polymeric substances (EPSs) play a major role in formation of biofouling in membrane processes [9]. EPSs consist of a variety of organic substances including polysaccharides, necleic acids, and proteins. Their major functions are to form a protective layer for the cells against the harmful external environments and to provide the sticky sturcture of the biofilm [10]. Therefore, bacteria embedded in the biofilm consisting of EPSs are more resistant to disinfectant. Many studies have reported the effect of EPSs in membrane bioreactors treating wastewater. However, few studies on biofouling of RO membranes have been performed due to relatively low concentration of EPSs on the process. Therefore, the main objectives of this research were to investigate bacterial adhesion or biofouling formation potential on RO membrane surfaces under differnt physico-chemical conditions such as ionic strength and to determine the effect of chlorine addition on EPS and biofouling reduction.

2. Methods and materials

2.1. RO membrane and feed water sources

The RO membrane (RE 1812-60, Seahan Industry) used in this study was a thin film composite polyamide membrane. Two different water sources were used to compare the effects of water properties on biofouling: (1) conventionally pretreated river water with relatively low organic content and low ionic strength, and (2) secondary effluent with relatively high organic content and high ionic strength. The conventionally treated water was alum coagulated, settled, and sand filtered Han River water (Seoul, Korea). The secondary effluent was collected from a sewage treatment plant (Seoul, Korea). Key parameters of water qualities of feed water sources are listed in Table 1.

2.2. Biofilm formation potential under a dynamic condition

An experimental system was installed to observe biofilm formation potential of two feed water sources as shown in Fig. 1. The system was designed to simulate a cross-flow type of membrane operation without permeate drag. The system consisted of two columns with a length of 50 cm and an inner diameter of 2 cm. Each column contained ten ring-shaped glass or membrane coupons. Each of the glass coupons (length 2.5 cm, OD 2.0 cm, and ID 1.8 cm) has a total surface area exposed to water of 12.6 cm^2. The membrane coupons were also made with the same size of the inner surface as the glass coupon. The column was continuously operated at a flowrate of 23 L/d. The feed water in a 20-L reservoir was changed every 3 days to maintain reasonably constant water qualities. The coupons were periodically withdrawn from the columns for further analyses including viable cell counts, EPSs, dried weights, and volatile suspended solids (VSSs).

2.3. Membrane filtration experiments

A series of membrane filtration experiments were performed to investigate how biofouling on membrane surfaces directly affected a permeate flux decline. A stirred batch cell (Millipore, USA) was operated at a constant pressure of 60 psi using nitrogen gas. An electronic balance (AND GF-2000, USA) was used to

Table 1
Characteristics of feed water sources in this study.

Parameter	Pretreated water	Secondary effluent
pH	7.45±0.2	7.32±0.3
Turbidity (NTU)	0.27±0.2	1.15±1
UV_{254} (cm^{-1})	0.019 ± 0.005	0.122±0.04
TOC (mg/L)	1.14±0.5	5.8±1.2
DOC (mg/L)	1.05±0.05	5.2±1.2
Viable cell counts (CFU/mL)	1.2×10^{-3}–1.3×10^{4}	1.3×10^{5}–1.5×10^{6}
Biodegradable organic carbon (mg/L)	0.4±0.08	0.6±0.1
Conductivity (μS/cm)	149.9±20	555.5±50

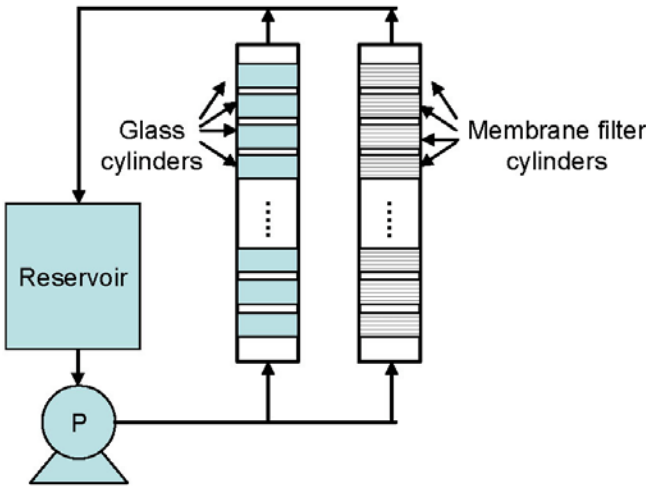

Fig. 1. Schematic of the experimental system for the biofilm formation potential test.

continuously measure permeate mass. After the membrane filtration experiments, further analyses of the membrane surface were conducted including viable cell concentrations, EPS, dried weights, and VSSs. In the chlorine addition experiment, feed water sources were treated with sodium hypochlorite (2 or 5 ppm) for 30 min and dechlorinated with 0.1 N $Na_2S_2O_3$ prior to biofouling formation tests.

2.4. Analytical methods

Biofilm samples were periodically collected from the surfaces of the materials and analyzed for dry weights, VSSs, viable cell counts, and EPSs. The dry weight and VSSs were measured according to gravitational measurements in Standard Methods [11]. For viable cell counts, the biofilm sample was homogenized and serially diluted in test tubes with sterile saline solution (3.5 g/L of KH_2PO_4, 4.3 g/L of K_2HPO_4, 8.5 g/L of NaCl). One-tenth mL of each dilution was spread in triplicate on R2A agar (Difco, USA). After spreading, the R2A plates were incubated at room temperature. The colony forming units on the agar plates were calculated by averaging colony numbers in triplicate samples after a 5-day incubation period. The samples for EPS analysis were extracted with EDTA (2%; at 4°C for 3 h) followed by high-speed centrifugation (20,000 g) for 20 min. A 0.2 μm membrane filter was used to remove microbial cells and a dialysis membrane (Cellu Sep, 3500 Da) was used to remove low molecular-weight metabolites (4°C for 24 h). The total quantity of extracted EPS was measured by total organic carbon (TOC).

The biodegradable organic carbon (BDOC) of the liquid samples was determined by the difference in DOC concentrations before and after 7 days of incubation period. The BDOC was used as an indicator of biofilm formation potential. Other analyses were followed by Standard Methods [11].

3. Results and discussion

3.1. Biofilm formation potentials with different feed waters

Two feed water sources, i.e., the pretreated water and the secondary effluents, were selected based on different characteristics such as organic contents and ionic strength. The biofilm formation potential test was performed using two different materials, glass and RO membrane, to understand effects of surface properties on biofilm formation potential. The results of viable cell concentrations are shown in Fig. 2. The surfaces exposed to the secondary effluent showed higher bacterial concentrations than those exposed to the pretreated water. The higher biofilm formation on the surfaces contacted with the secondary effluents was primarily due to the greater carbon sources (i.e., higher TOC and DOC) and the higher ionic strength (i.e., higher conduictivity). In addition, viable cell concentrations on the membrane surface were higher than those of the glass surface. The roughness and pore geometry of membrane filters were known to have impacts on cell attachments on membrane surfaces [4]. The result indicated that the membrane surface was preferable to microbial adhesion and thus to biofouling compared to the glass surface.

3.2. Characteristics of deposition on the fouled membrane surfaces

Effects of biofouling on membrane performance were observed through a batch test of membrane filtration. During the test, permeate was measured and calculated as membrane flux. The membrane flux using the secondary effluent showed a more rapid decline than that using the pretreated water, indicating higher fouling tendency of the secondary effluent as anticipated (Fig. 3).

When the permeate flux dropped to 40% of the initial value, the membrane filtration was stopped and the biofilm samples on the membrane surface was collected for the analyses of biofoulant accumulation in terms of viable cell counts, dry weights, VSSs, and EPSs as shown in Table 2. The higher bacterial concentration on the membrane surface was observed when the secondary effluent was subjected to the membrane process, although the tested time was short and the amount of the permeate was smaller than that of the pretreated water. To normalize with produced water volume, the cell concentrations were divided with permeate throughput volume. The result showed that the attached cells on the membrane surface with the secondary effluent were

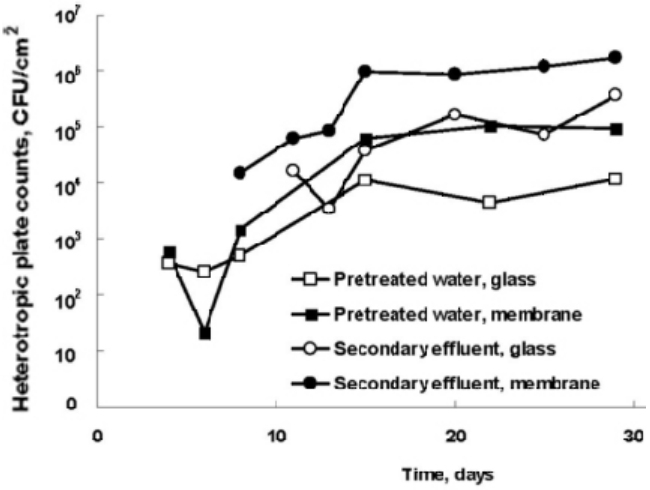

Fig. 2. Viable cell concentrations accumulated on the surfaces of glass and membrane.

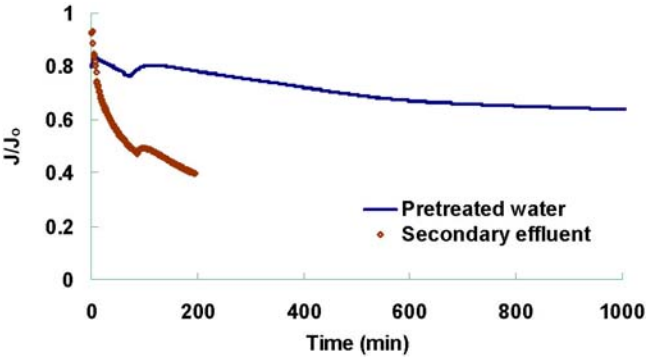

Fig. 3. Flux decline of the pretreated water and secondary effluent.

approximately three orders of magnitude higher than those with the pretreated water.

The mass of total solids normalized with the produced permeate volume were similar in both pretreated water and secondary effluent. The ratios of the VSSs to the dry weights were approximately 10% for the pretreated water and 20% for the secondary effluent. In addition, the EPS concentration consisted of almost 98% of the dry weight for the secondary effluent, whereas it was approximately 6% of the dry weight for the pretrated water. These results implied that the flux decline of the membrane exposed to the secondary effluents were mostly due to the biofilm formation consisting mainly of EPSs.

3.3. Effects of chlorine addition on biofilm control

Since chlorine disinfection has been widely used to control biological activities in RO membrane processes, in this study, effects of chlorine addition on biofouling control were investigated. The feed water sources were chlorinated at a selected dose and then were dechlorinated. The membrane filtration test were performed and then the membrane surfaces were analyzed. The results of membrane filtration are shown in Fig. 4. The membrane

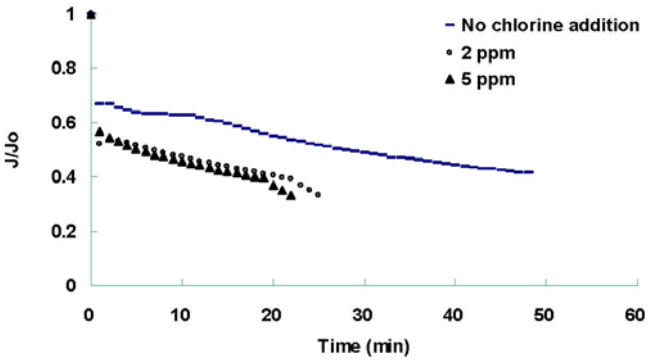

Fig. 4. Flux decline of secondary effluent with chlorination.

Table 2
Characteristics of foulant on the surfaces after membrane filtration

	Pretreated water	Secondary effluent
Normalized with surface area of the membrane:		
Viable cells, CFU/cm^2	4.3×10^4	1.5×10^6
Dry weight, mg/cm^2	0.612	0.05
VSS, mg/cm^2	0.064	0.01
EPS, mg/cm^2	0.034	0.048
Normalized with surface area of the membrane and permeate volume:		
Viable cells, CFU/cm^2	5.7×10^4	2.2×10^7
Dry weight, mg/cm^2	0.81	0.71
VSS, mg/cm^2	0.084	0.141
EPS, mg/cm^2	0.045	0.674

Table 3
Feed water qualities and EPS concentrations of the biofouled membrane during the chlorination tests

Parameters	Pretreated water	Secondary effluents	
	Cl_2, 2 mg/L	Cl_2, mg/L	Cl_2, 5 mg/L
UV_{254} (cm^{-1})	0.04±0.015	0.15±0.02	0.17°0.02
TOC (mg/L)	1.65±0.5	6.87±1.0	6.25±1.3
Conductivity (μm/cm)	226.8±20	648.0±60	707.3±60
Viable cell counts (CFU/mL)	6×10^2	4×10^4–7×10^5	2×10^4–5.6×10^5
EPS, C/Co^a	0.81	1.2	1.2

performance deteriorated with chlorine treatment although the viable cell concentrations were generally decreased after chlorination as listed in Table 3. The viable cell count without chlorination was approximately ~108 values, but it decreased approximately three or four orders of magnitude when chlorination was applied. This finding indicated that microorganisms was inactivated by chlorination although the EPS concentration was not decreased. Consequently, chlorination was proven to be ineffective to biofilm detachment or to reduction of biofilm adhesion.

4. Conclusions

To successfully apply RO membrane processes for reclamation and reuse of municipal wastewater effluents, a suitable pretreatment process should be studied and suggested to overcome the biofouling problem. Investigation of biofouling formation potential was conducted using wastewater effluents and conventionally pretreated water. As expected, bacterial adhesion on the surface of the membrane was greater in wastewater effluents due to preferable physicochemical interactions of microbial cells and membrane including high ionic strength and roughness. The membrane biofouling was detected with a flux decline pattern and analyses of the fouled membrane. Especially, the results of EPS concentrations implied that biofouling was the main cause of membrane fouling. The common biofouling control method, i.e., chlorination, should be used with caution because the flux decline was not improved with the chlorination step. In addition, the chlorination was ineffective in reducing the EPS concentrations on the membrane surface, and thus failed to enhance bacterial detachment.

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