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The role of physical cleaning conditions and coagulation on algal-rich water fouling of the ceramic membrane

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ABSTRACT

In this paper, we tried to derive the membrane fouling characteristics of *Chlorella*, green algae, under various operating conditions by using a ceramic membrane. To this end, this study examined the effects of fouling on ceramic membranes according to flux and coagulant and the effects of *Chlorella* attached to the membrane surface due to backwash (B/W) in filtering ceramic membranes. Therefore, we tried to derive the relationship between B/W and membrane fouling control under various conditions by analyzing the membrane fouling phenomenon through filtration resistance analysis according to each operation condition. When filtering membranes in algal-rich water, the selection of coagulants is an important factor. It was confirmed that aluminum-based coagulants were more effective in membrane fouling control than iron-based coagulants. When filtering membranes in algal-rich water, the selection of coagulants is an important factor. It was confirmed that aluminum-based coagulants were more effective in membrane fouling control than iron-based coagulants. This study examined the control properties of algal membrane fouling depending on various B/W conditions (strength, time, and temperature). It was confirmed that physical cleaning efficiency was found only by ensuring B/W beyond a certain period. Furthermore, it was confirmed that B/W strength was more effective in fouling control than time duration beyond a certain period. The efficiency depending on mitigating fouling layers on the membrane fouling with an increasing water temperature of B/W.

Keywords: Algae; Membrane fouling; Physical cleaning; Coagulation

1. Introduction

Algae are not only important food sources for aquatic organisms such as fish and crustaceans but also important creatures that produce oxygen using photosynthetic by-products to support aerobic organisms in aquatic systems. However, global warming has brought changes in a water environment thus far. In particular, the occurrence of algal blooms causes serious problems (e.g., disinfection by-products, unpleasant taste and odor) in water supply due to an increase in water temperature during summer in water environments such as rivers or lakes and then eventually causes undrinkable water. Recently, low-pressure membrane filtration such as ultrafiltration and microfiltration (MF) has attracted increasing attention from algal rich water treatment areas because of the higher production of treated water and lower operating costs. According to preliminary studies [1–3], a membrane process was able to completely remove algal cells, and a hybrid process combined with activated carbon was able to remove extracellular organic matter (EOM). Lately, the development of ceramic membrane manufacturing technologies has allowed becoming as competitive as polymeric membranes, and then they have received attention from the water treatment field. Recently, the excellent

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chemical resistance of ceramic membranes makes it possible to solve problems related to periodic repair and replacement as a disadvantage of polymeric membranes. However, little is known about the fundamental properties of ceramic membrane fouling caused by algal materials. Ceramic membrane processes have rapidly gained attention because of many more specific advantages compared to existing polymeric membrane processes.

Ceramic membranes have been applied to drinking water treatment for 50 facilities in Japan in the last decade. Recently, a 2.5 mega gallons per day pilot plant has been studied in California, the United States [4,5].

Ceramic membranes have been widely applied in water treatment, but membrane fouling remains an important issue until membrane technologies are widely applied. Also, little is known about their performance properties, particularly membrane fouling caused by algae.

It has been reported that membrane fouling in applying algae-rich water to membrane processes is mainly caused by algal cells including similar substances such as polysaccharides, proteins, lipids, and humus and EOM [6]. The forms of membrane fouling include adsorption or gelation between algal particles and the membrane surface and formation of pore blockage or cake layers [7].

Many studies have been conducted as a method of controlling membrane fouling. Recently, the processes combined with coagulation, adsorption, and oxidation processes have been widely studied to control membrane fouling [8], and it was reported that backwash (B/W) and air scrubbing (A/S) were effective alternatives to improve membrane performance [9].

In this paper, we tried to derive the membrane fouling characteristics of *Chlorella*, green algae, under various operating conditions by using a ceramic membrane. To this end, this study examined the effects of fouling on ceramic membranes according to flux and coagulant and the effects of *Chlorella* attached to the membrane surface due to B/W in filtering ceramic membranes. Therefore, we tried to derive the relationship between B/W and membrane fouling control under various conditions by analyzing the membrane fouling phenomenon through filtration resistance analysis according to each operation condition.

2. Materials and methods

2.1. Characteristics of the raw water

The single culture samples of *Chlorella* algae were collected from Danyang *Chlorella* in South Korea. To understand fouling by membrane material, the *Chlorella* concentration was adjusted to 100 mg/L (3.98 g/m^2 of algal deposition) suspended solids as shown in Table 1.

2.2. Experimental set-up

All the experiments were conducted using a laboratory-scale membrane set-up, mainly including a raw water tank, a constant level water tank, a peristaltic pump, a pressure transducer, and a data acquisition system. As shown in Table 2, the immersed flat type MF membrane module (Cembrane, Denmark) was made of silicon carbide, with an effective membrane area of 0.0652 m^2 and a nominal pore size of 0.1 µm. The reactor (effective volume of 16.4 L) was fed with a raw solution through the constant level tank and the effluent was drawn directly from the membrane module by the peristaltic pump (EMS-2000S, Korea). A pressure transducer (PTP708 Tuopu Electric, Korea) was connected to a laptop, was used to continuously monitor the transmembrane pressure (TMP).

2.3. Operating conditions

Through the preliminary test, the critical flux and SADM (specific aeration demand, either to the membrane area) of the ceramic membrane used in this study were 200 LMH and 1 m^3/m^2 h, respectively. The flux and A/S rates were selected based on the above preliminary test results. An analysis of membrane fouling by coagulant type was performed by selecting $FeCl_3$ and polyaluminium chloride (PaCl). Also, it was conducted to examine the fouling properties according to the conditions of B/W under the following conditions: with/without B/W, with/without chemicals, temperatures (10°C, 20°C, and 30°C), and with/without A/S. The reactor temperature during the experiments was maintained at 20°C ± 0.5°C using a water bath, and each membrane was washed and flushed by ultrapure water under the same conditions before use.

2.4. Measurement of resistance

Analysis of membrane fouling by *Chlorella* was using the following equations according to the measurement of permeate flux, transmembrane pressure, and water temperature: In filtration experiments, the specific membrane resistance was first measured using ultrapure water.

Moreover, the reversibility and irreversibility under each condition were analyzed using the following methods: Membrane fouling was calculated by the following equations in defining the filtration resistance obtained by filtering ultrapure water through the initial membrane, the final filtration resistance of the fouled membrane by filtering using the target raw water to be treated, and the filtration resistance obtained by filtering using ultrapure water after physical cleaning as R_0, R_1, and R_2, respectively:

$$\mathrm{RF} = \frac{R_2 - R_1}{R_0 - R_1} \tag{1}$$

$$\mathrm{IF} = \frac{R_0 - R_2}{R_0 - R_1} \tag{2}$$

$$\mathrm{TF} = \mathrm{RF} + \mathrm{IF} = 1 \tag{3}$$

RF is reversible fouling, IF is irreversible fouling and the total membrane (TF) becomes 1.

3. Results and discussion

3.1. Effects of TMP with injection of coagulants

Fig. 1 shows the fouling properties with and without injecting coagulants and by coagulant type. The injection volume of $FeCl_3$ and PaCl were determined by jar-test

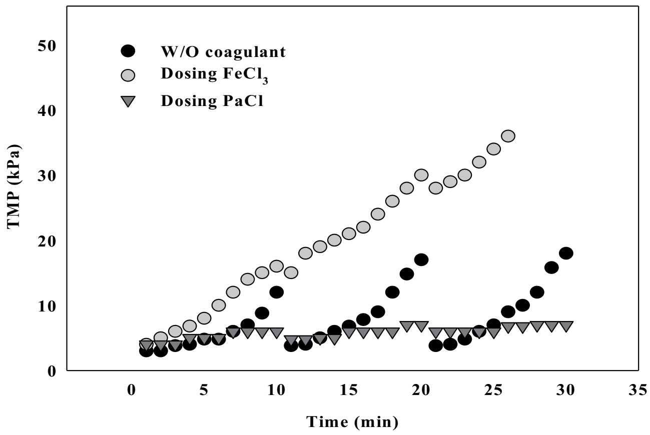


Fig. 1. Comparison of operating TMP by coagulant type and with/without injection of coagulants.

at 12.5 and 7.0 mg/L, respectively. As shown in the figure, the rates of increase in TMP were dosing $FeCl_3$, without (W/O) coagulant, and dosing PaCl, in that order. Coagulation conditions such as type, dose, and mix mode of coagulants have a large effect on coagulation-membrane processes [10,11]. Additionally, Konieczny et al. [12] reported that aluminum salt coagulant showed higher removal efficiency than ferric salt. The TMP increase rate of iron-based coagulants was higher than that of aluminum-based coagulants also in this study. In particular, the TMP increase rate with the dosage of iron-based coagulants was higher than without them. Moreover, the PaCl dosage maintained lower TMP during the filtration time than W/O. It is considered that these results with the PaCl dosage were obtained by coagulating foulants into large aggregates to improve the water permeability. According to previous literature, it has been reported that fouling is caused by algae cells and EOM including polysaccharides proteins, lipids and humus [6], and it is known that membrane fouling decreases with increasing coagulant dosage into these substances by coagulating them into large aggregates [13]. However, the PaCl dosage showed a higher increase in initial filtration pressure than W/O after the third cycle. Foulants connected to aluminum ions showed increased adsorption performance with the membrane surface. Wu et al. [6] reported that the injection of coagulants accelerated fouling by causing low electrostatic repulsion between particles and membrane surface due to an increase in the zeta potential of feed water. Similar results were obtained also in this paper.

3.2. Fouling control properties depending on A/S shear force in B/W sequence

This study aimed to examine the effects of fouling control according to physical cleaning conditions in membrane filtration, that is to say, with or without A/S and B/W as shown in Fig. 2. It was confirmed that A/S showed larger effects of fouling control than B/W when filtering membranes in algal-rich water. The experiments were performed to examine the effects of A/S on fouling control in B/W sequence, and without A/S, 0.5 m/h (SAD_m), and 1.0 m/h (SAD_m) was selected as an operating condition. As shown in Fig. 3, A/S

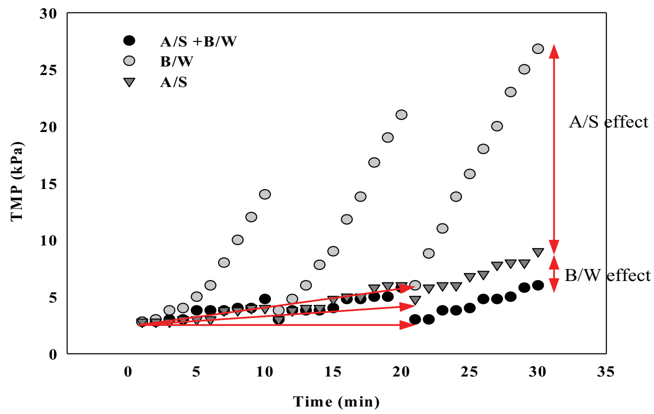


Fig. 2. Fouling properties with/without A/S and B/W.

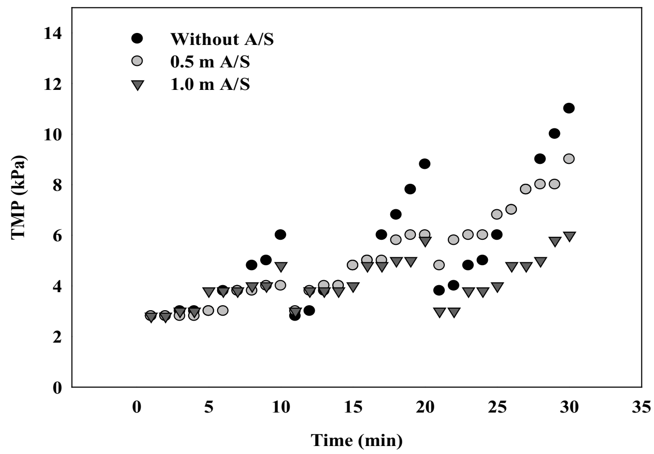


Fig. 3. Effects of A/S on B/W sequence.

detaches algae foulants attached to the membrane surface during the B/W process. According to previous literature, extracellular polymeric substances (EPS's) adhesive and cohesive forces released from algal cells are not covalent bond forces but physicochemical forces. It has been reported that this EPS makes it easy to remove membrane fouling caused by cellular adhesion but cells are difficult to be removed [14]. Also, it has been reported that it is very hard to remove algal cells because they are many more than the total number of bonds and covalent bonds even though they have disadvantages compared to C–C bonds and algal cells are irreversibly attached even though filtration processes stop [15]. Interactions with the membrane surface made it easy for algal cells to be adsorbed also in this study. It is regarded as intermediate fouling, and this type of fouling shows higher physical cleaning efficiency with increasing shear force of A/S.

3.3. Fouling control properties depending on B/W strength and operating conditions

The experiments were performed with different B/W strength (1.5, 2.0, and 3.0 Q, 30 s) to examine the effects of B/W strength on membrane fouling control. Fig. 4 shows that the TMP increase rate is similar during the initial pressure

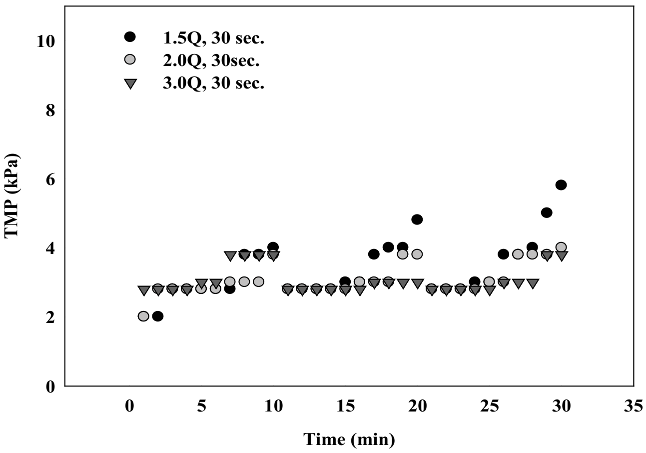


Fig. 4. Comparison of TMP depending on B/W strength.

Table 1
Characteristics of the raw water

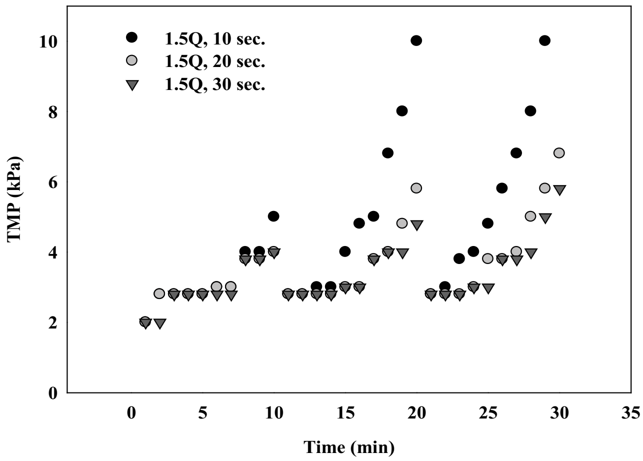
Parameters		Concentrations
Chl-a		1,460 ± 23 mg/m^3
Cell		3.98 ± 1,2 g/m^2
Suspended solids		100 ± 12 mg/L
EPS	Protein	0.782 ± 0.5 mg/L
	Polysaccharide	8.9 ± 1.2 mg/L

Table 2
Physical characteristics of membrane

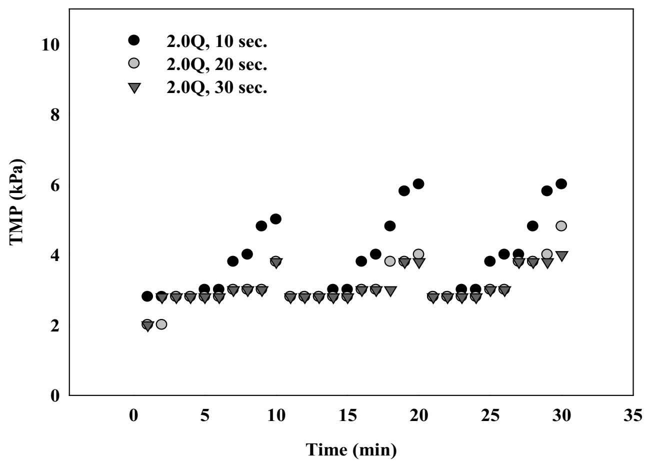
Characteristic items	Properties
Membrane material	Silicon carbide
Effective filtration area (m^2)	0.00652
Membrane type	Flat type
Pores if the MF membrane (μm)	0.1
Clean water permeability (LMH/bar)	5,000 LMH/bar at 20°C

recovery and filtration time at B/W of more than 2.0 Q. The experiments were conducted to investigate the physical cleaning efficiency with changing B/W time (10, 20, 30 s) and B/W strength conditions (1.5, 2.0, 3.0 Q). As shown in Fig. 5, the TMP increase rate is similar at B/W of more than 2.0 Q and for more than 20 s, and more stable TMP was found at B/W of 3.0 Q during the filtration. These experiments showed that B/W time and strength affected detaching algal fouling as a B/W condition. B/W strength had an intense effect on fouling control beyond a certain period (20 s).

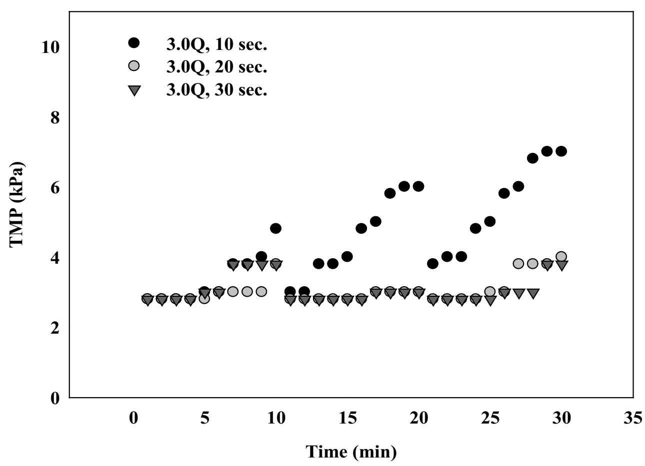
The following experiments were performed to examine the membrane fouling control properties depending on B/W temperatures by changing the B/W conditions 10°C, 20°C, and 30°C at B/W of 2.0 Q for 30 s. Fig. 6a shows that the stability of the operating results increased with increasing B/W temperature. Moreover, Fig. 6b shows the formation of membrane fouling resistance depending on the above B/W conditions. Rir resistance tended to decrease with increasing B/W temperature. Ren et al. [16] reported that B/W substantially mitigated fouling layers formed in membranes at a higher temperature and its cleaning efficiency increased at a higher temperature. Similar results were also found in this study. 3.3



(a)

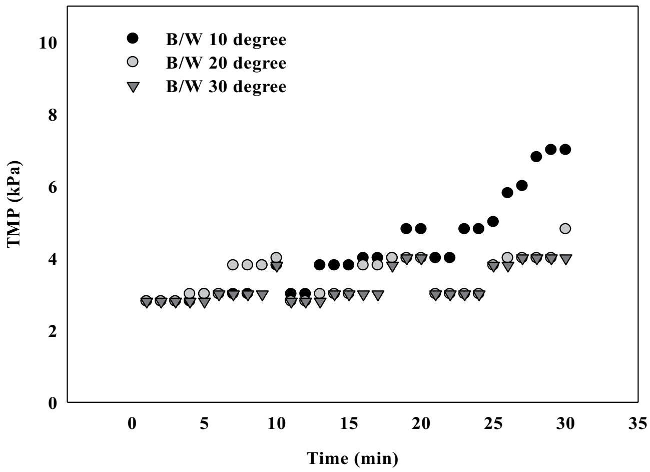


(b)

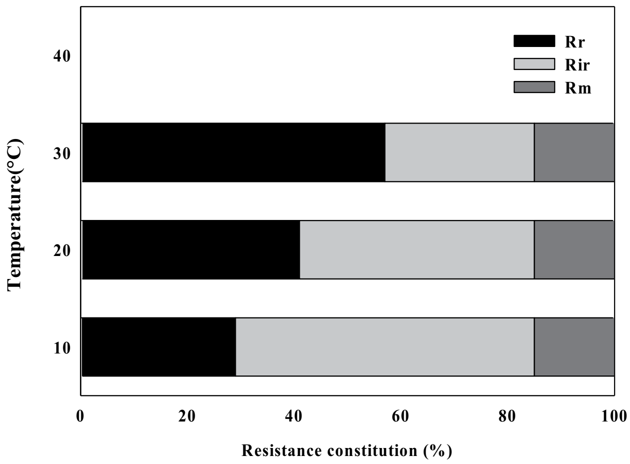


(c)

Fig. 5. B/W strength and time to TMP comparison (a) B/W 1.5Q, (b) B/W 2.0Q, and (c) B/W 3.0Q.



(a)



(b)

Fig. 6. (a) Compare of operating TMP and (b) membrane fouling resistance at different backwashing temperatures.

made it possible to examine the membrane fouling control properties depending on strength, duration, and temperature among various B/W conditions.

4. Conclusions

In this paper, we tried to derive the membrane fouling characteristics of *Chlorella*, green algae, under various operating conditions by using a ceramic membrane. To this end, this study examined the effects of fouling on ceramic membranes according to flux and coagulant and the effects of *Chlorella* attached to the membrane surface due to B/W in filtering ceramic membranes. Therefore, we tried to derive the relationship between B/W and membrane fouling control under various conditions by analyzing the membrane fouling phenomenon through filtration resistance analysis according to each operation condition. The following conclusions were drawn from this study.

- When filtering membranes in algal-rich water, the selection of coagulants is an important factor. It was confirmed that aluminum-based coagulants were more effective in membrane fouling control than iron-based coagulants. Furthermore, membrane fouling was worse than without injecting coagulants when selecting $FeCl_3$. It is considered that the selection of coagulants is an important factor when concentrating or filtering with *Chlorella* membrane processes.
- A/S is an important element in membrane fouling control when filtering *Chlorella*. It was confirmed that A/S is an effective method for controlling deposited algal foulants. Moreover, A/S had a large effect on detaching algal foulants attached to the membrane surface in B/W. It was confirmed that higher physical cleaning efficiency increased with increasing shear force of A/S.
- This study examined the control properties of algal membrane fouling depending on various B/W conditions (strength, time, and temperature). It was confirmed that physical cleaning efficiency was found only by ensuring B/W beyond a certain period. Furthermore, it was confirmed that B/W strength was more effective in fouling control than time duration beyond a certain period. The efficiency depending on mitigating fouling layers on the membrane fouling with an increasing water temperature of B/W.

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