Recovery of filtration performance by steam cleaning with a low NaOCl concentration in a ceramic membrane process

Sun-A An[a], Joon-Seok Kang[b], Jeong-Eun Lee[a], Soyeon Kang[a], Han-Seung Kim[a,*]

[a]*Department of Environmental Engineering and Energy, Myongji University, 116 Myongji-ro, Cheoin-gu, Yongin-si, Gyeonggi-do 17058, 449-728, Korea, Tel. +82-031-330-6695; emails: kimhs210@mju.ac.kr (H.-S. Kim), suna9602@naver.com (S.-A. An), leeje1610@gmail.com (J.-E. Lee), tkfdk3743@naver.com (S. Kang)*
[b]*Department of Water Resource, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Korea, email: joonseok0724@gmail.com (J.-S. Kang)*

ABSTRACT

In the membrane separation process, a decrease of filtration performance is caused by contaminants adsorbed on the pores of the membrane. Therefore, membrane cleaning is carried out in order to recover the membrane performance. Irreversible fouling that cannot be eliminated by physical backwashing is treated with chemicals. While in the case of organic polymer membranes, high concentration of chemicals facilitates the aging of the membrane, and restricts its cleaning condition. In this study, to compare the cleaning recovery rate against the fouled membrane using the ceramic membrane which has chemical resistance, the operating pressures were set at 100, 200, and 300 kPa and the raw water contained the turbidity of 10 and 25 NTU, and the dissolved organic matter (DOC) of 2.5 and 8 ppm. The higher the operating pressure and the DOC concentration were set, the higher the filtration flux reduction rate generated. Three cleaning methods were executed for the fouled membranes: physical backwashing, steam cleaning, and chemically enhanced steam cleaning (CESC). The cleaning methods including steam expressed a better cleaning recovery rate than the physical backwashing. CESC used sodium hypochlorite (NaOCl) as detergent not only indicated a high cleaning recovery rate at the beginning of cleaning but also improved cleaning recovery over time, unlike steam cleaning that has a constant value in recovery rate despite the longer cleaning time. It is supported that NaOCl steam cleaning is suitable as a cleaning method to complement physical backwashing.

Keywords: Ceramic membrane; Membrane performance recovery; NaOCl; Steam cleaning; Chemically enhanced steam cleaning

1. Introduction

The importance of water treatment technology is emerging as a global water shortage phenomenon. The advantages of the membrane separation process have been more emphasized because the site area can be minimized, and the water quality can be secured rather than sand filtration process. The separation membrane is divided into an organic membrane and an inorganic membrane according to the material. In the membrane separation process, inorganic particles and dissolved organic matter (DOC) in raw water cause membrane fouling, resulting in the reduction of permeability and production quantity [1]. For contaminated membrane, physical backwashing and cleaning in place (CIP) are typically executed to recover the filtration performance. Physical backwashing by using a high-pressure DI (deionized water) and air is widely applied than CIP. The cycle of CIP is considered by the stain resistance,

* Corresponding author.

membrane fouling morphology, and economic efficiency when membrane fouling that cannot be recovered by physical backwashing occurs. It requires a high concentration of chemicals and long cleaning time. Chemical enhanced backwashing (CEB) is carried out when the membrane performance deteriorates applying relatively low concentration of chemicals and short cleaning time. Frequent performance of CIP and CEB causes problems such as lowered total production quantity, high cost of chemicals and waste water treatment [2,3]. Sodium hypochlorite (NaOCl), one of the cleaning agents used to remove organic substances deposited on the membrane, is dissociated into water and produces reaction intermediates (ClO, OH radicals). An excessive dosage of NaOCl, however, damage the organic polymer membrane, consequently, the membrane lifetime is shortened [4,5]. Organic polymer membrane is not only low in chemical stability but also damages the membrane via high pressure and high temperature, so it is impossible to operate in extreme conditions and has limitations in membrane cleaning. On the other hand, the ceramic membrane that is not influenced by pH has remarkable physical and chemical stability, so it can be used semi-permanently through various cleaning methods [6]. Also, the ceramic membrane has occurred much less irreversible fouling because their interaction with foulants was weaker than polymeric membranes [7]. To minimize irreversible fouling, studies have been conducted to reduce the surface hydrophobicity by improving water contact angle using alumina sol [8]. In case of monolith ceramic membrane, pretreatment processes such as flocculation and sedimentation are not demanded via its characteristics that promote re-aggregation despite short hydraulic residence time [9]. Recently, steam cleaning technology has been reported to reduce the frequency of CIP and maximize operating efficiency. The high thermal energy of steam breaks the bond between the membrane and contaminants through pyrolysis (Fig. 1). It has concluded that steam cleaning was effective in controlling the sudden membrane contamination of organic matter [10]. In this study, it is aimed to identify membrane fouling under extreme conditions such as high concentrations of natural organic matter (NOM) and high pressure operating and to improve the recovery rate though new cleaning method by contacting steam mixed with sodium hypochlorite of low

concentration to the ceramic membrane. It is defined by chemically enhanced steam cleaning (CESC) in this article. It is possible to improve the operating efficiency of water purification plants by reducing CIP frequency. We evaluate the applicability of the new cleaning method by comparing the efficiency of steam cleaning with and without NaOCl.

2. Materials and methods

2.1. Ceramic membrane microfiltration process

The schematic diagram of ceramic membrane filtration is displayed in Fig. 1. Filtration was conducted at 100, 200, and 300 kPa. Turbidity and DOC concentration as main causes of membrane fouling were set as case 1 for 10 NTU of turbidity and 2.5 ppm of DOC based on the average value of raw water of water purification plants in Seoul from '09–'17 [11,12]. Based on this, membrane fouling characteristics and the membrane performance recovery according to the cleaning methods were experimented at high turbidity (25 NTU) and high DOC concentration (8 ppm). The raw water characteristics are exhibited in Table 1. Kaolin (SHOWA Chemical, Gyoda, Japan) was induced by turbidity. Humic acid (Sigma Aldrich, St. Louis, MO, USA) of 1,000 mg/L, which had been stirred for 48 h, was filtered through 1.2 μm glass microfiber filter (GF/C 0.47, Whatman, Maidstone, UK) and the solution was used to set the DOC. Humic acid and fulvic acid are typical examples of hydrophobic organic substances. It is known that humic acid has more molecular weight and higher hydrophobicity tendency than fulvic acid [13]. In the experiment, humic acid was used to

Table 1
Characteristics of raw water

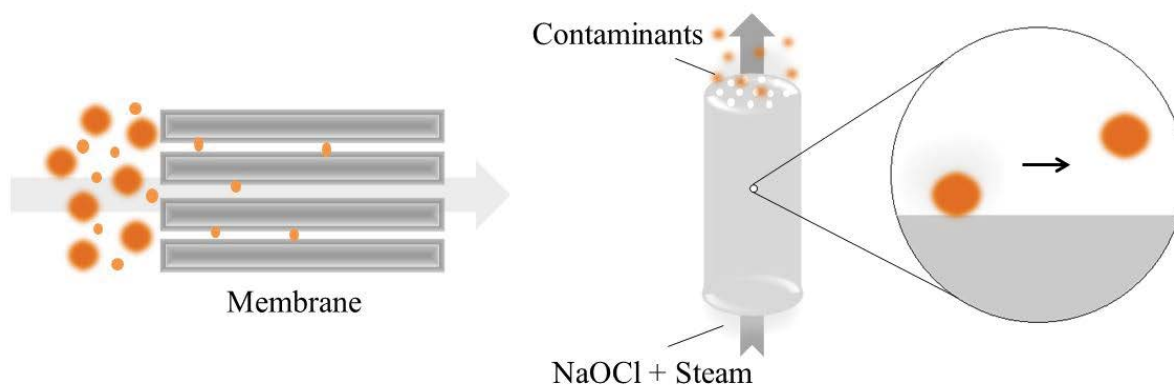|	Turbidity	DOC concentration
Case 1	10 NTU	2.5 mg/L
Case 2	25 NTU	2.5 mg/L
Case 3	10 NTU	8 mg/L
Case 4	25 NTU	8 mg/L

Fig. 1. Schematic diagram of steam cleaning method with NaOCl.

confirm the cleaning effect of the fouled ceramic membrane due to excessive hydrophobic adsorption.

The MF membrane was made of Al_2O_3 as a ceramic membrane of METAWATER Co., (Tokyo, Japan). One ceramic membrane module that has a pore size of 0.1 μm and a membrane area of 0.035 m^2 was used. Details of the membrane and figure of the module are shown in Table 2.

2.2. Experimental procedure and analytical methods

Ceramic membrane filtration was carried out by dead-end filtration and constant pressure filtration of 100, 200, and 300 kPa. Flux was calculated by converting an electronic balance (FG-60KAL-H, A&D Company, Tokyo, Japan) into a volume through the weight of the effluent increased for 10 s with a computer program. The degree of restoration of the membrane was evaluated though the following equation.

$$\text{Membrane recovery rate}(\%): \frac{J_2 - J_1}{J_0 - J_1} \times 100 \tag{1}$$

where J_0 is the pure water flux before filtering raw water; J_1 is the final flux of the fouled membrane before cleaning; J_2 is the pure water flux of the fouled membrane after cleaning.

The filtration end point was set to be about twice the volume of the ratio of flux (J/J_0) below 20% at 25 NTU of turbidity and 300 kPa of operation pressure. Raw water for DOC of 2.5 ppm was filtered 15 L and raw water for DOC of 8 ppm was filtered 2.5 L. J_0 and J_2 have adopted a value that the flux is kept for 1 min when DI was filtered. Water quality analysis of influent and effluent was carried out in order to check the pollutant removal ability by membrane filtration. Turbidity, DOC, UV_{254}, temperature, pH, and Flux were measured. For DOC measurement, the sample was filtered with a 0.45 μm PP syringe filter (GD/X Syringe Filter, Whatman). The analytical instruments and method for each analysis are displayed in Table 3.

2.3. Cleaning methods

Physical backwashing (*B*) is performed by pressurizing 500 kPa of air for 1 s and backwashing for 3 s with DI of 500 kPa to take off contaminants during filtration. It takes about 1 min. To initialize ceramic membrane performance, after the end of the experiment the membrane was immersed with citric acid of 10,000 ppm and NaOCl of 3,000 ppm for 6 h at least, respectively. The ceramic membrane that was initialized by chemicals was stored in DI. Steam cleaning (SC) is a cleaning method in which 120°C of DI generated by the steam generator is contacted with the membrane to break the bond between the membrane and the contaminant. The pressure and temperature of the steam generator were maintained at 130–150 kPa and 118°C–124°C. Steam cleaning methods proceeded from 1 to 5 min. After cleaning with steam, physical backwashing facilitated the detachment of contaminants. CESC was injected into 1 mL of NaOCl at 510 ppm before steam discharged from the steam generator reached the membrane. Considering that the flow rate of the steam is 50 mL/min, NaOCl of 10 ppm contacted the membrane. All cleaning method experiments included steam were conducted after physical backwashing. Therefore, the physical backwashing was performed twice in all raw properties and the average value of there was used. The cleaning efficiency of steam cleaning and CESC used the average value of the flux measured for 1 min.

2.4. Resistance in series model

Membrane fouling can be interpreted quantitatively through resistance in series model (RISM). This model expresses the main factor of flux degradation as the sum of the resistances. The factors to be considered are membrane resistance (R_m) due to the membrane itself, cake layer resistance (R_c) formed on the surface of the membrane during filtration, and internal fouling resistance (R_f), which is caused by adhesion and adsorption of contaminants in membrane pores. This can be used to determine the type of membrane fouling and the degree of membrane recovery by cleaning. The relationship between each resistance value and flux (J), and trans-membrane pressure (TMP) can be shown as follows [14].

$$J = \frac{\text{Driving force}}{\sum \text{Resistance}} = \frac{\text{TMP}}{\mu \cdot R_T} \tag{2}$$

Table 2
Characteristics of ceramic membrane module

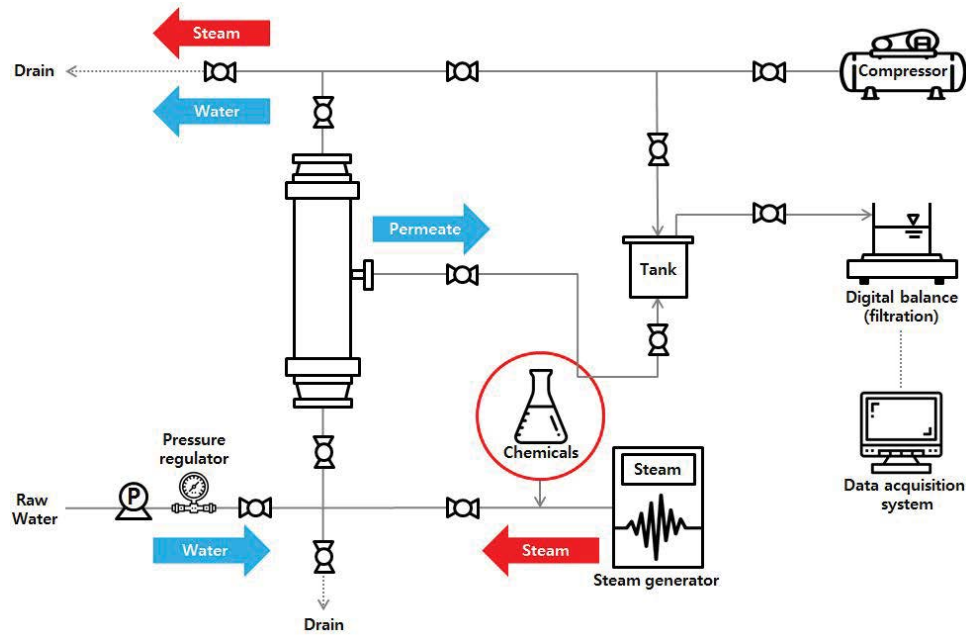| Ceramic membrane type	Contents	Membrane module
Material	Ceramic (Al_2O_3)	
Type	Inner-pressured type monolith	
Nominal pore size	0.1 μm	
Dimension	(Φ) 30 mm × 100 mm (L)	
Size of channel	f 2.0 mm	
Number of channels	55	
Membrane surface area	0.035 m^2	
pH range of acceptable	1–14	
Maximum operating pressure	1,961 kPa	
Manufactory	METAWATER (Tokyo, Japan)	

Fig. 2. Schematic diagram of a lab-scale system.

Table 3
Analytical instruments and methods

Categories	Analyzers	Etc.
Turbidity	2,100 N turbidimeter, HACH (Colorado, US)	NTU (nephelometric turbidity unit)
DOC	TOC-VCPH, Shimadzu (Kyoto, Japan)	NPOC (non-purgeable organic carbon)
UV_{254}	UV-1800, Shimadzu (Kyoto, Japan)	Ultraviolet photometer
Temperature	Orion 3star, Thermo (Massachusetts, US)	pH meter
pH	Orion 3star, Thermo (Massachusetts, US)	pH meter
Flux	FG-60KAL-H, A&D Company (Tokyo, Japan)	Weighing machine

$$R_T = R_m + R_c + R_f \tag{3}$$

$$J = \frac{TMP}{\mu\left(R_m + R_c + R_f\right)} \tag{4}$$

Flux (J), which represents the treated water quantity per unit membrane area per unit time, is proportional to the TMP as driving force on the membrane and is inversely proportional to the viscosity (μ) and the total resistance (R_T). The membrane fouling analysis through RISM could regard that the restoration of the flux implies reversible fouling by the cake layer resistance (R_c) and the non-restoration of the flux implies irreversible fouling by the internal fouling resistance (R_f) [15].

3. Results and discussion

3.1. Membrane fouling characteristics with water quality and operating pressure

Flux decline according to the pressure of case 1–4, which based on the turbidity and organic matter entering the water purification plants in Seoul, is shown in Fig. 3. The graph shows the change of J/J_0 (%) according to V/V_0 (%), where V/V_0 (%) is the ratio of the permeation volume. Total permeation volume of 2.5 and 8 ppm of DOC was 15 and 2.5 L, respectively. When comparing the results for DOC of 2.5 ppm (Figs. 3a and b) and DOC of 8 ppm (Figs. 3c and d), J/J_0 (%) is rapidly deteriorated by DOC rather than turbidity. It has been reported by Lee [16] that the DOC of raw water is composed of humic acid, which has a greater influence on the occurrence of membrane fouling than particles.

$$SUVA_{254}\left(L/mg\,m\right) = \frac{UV_{254}\left(cm^{-1}\right)}{DOC\left(\dfrac{mg}{L}\right)} \times \frac{100\,cm}{1\,m} \tag{5}$$

$SUVA_{254}$ which combined UV254 and DOC as one parameter enables to quantitative measurement of the aromatic content in the concentration of organic carbon. Particularly, the absorbance at the wavelength of 254 nm was measured because the interference by inorganic compounds can be minimized and the aromatic molecules have the largest
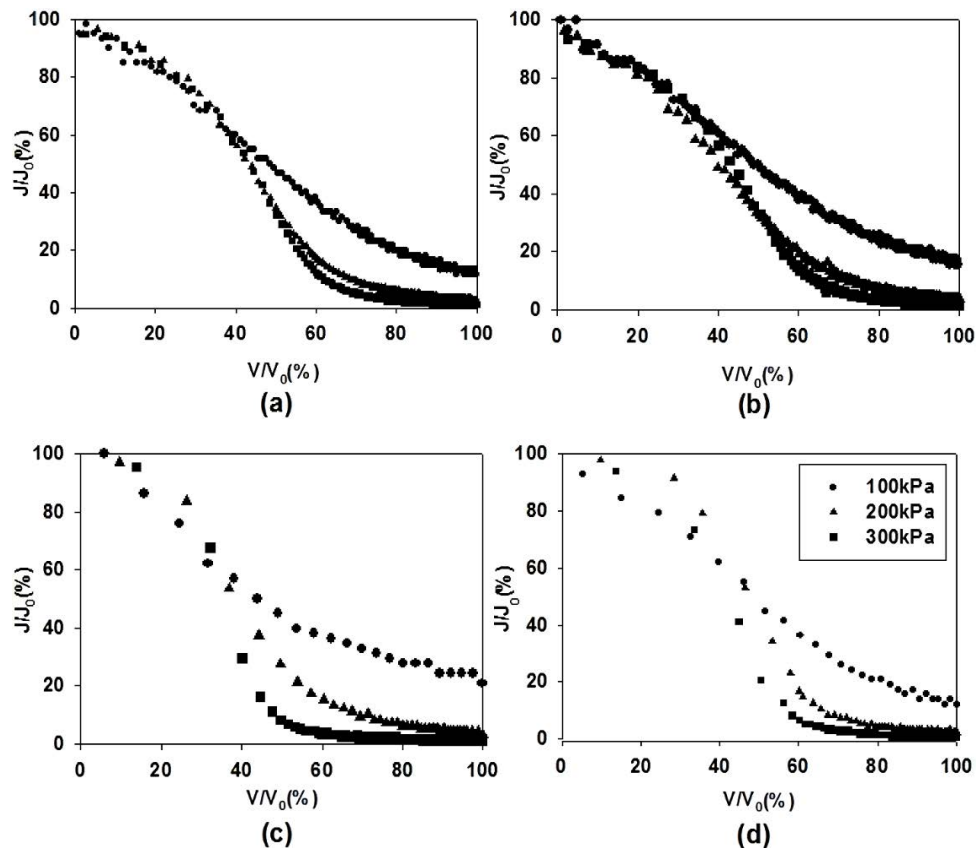
Fig. 3. Change of J/J_0 (%) according to V/V_0 (%): (a); 10 NTU of turbidity, 2.5 ppm of DOC, (b); 25 NTU of turbidity, 2.5 ppm of DOC, (c); 10 NTU of turbidity, 8 ppm of DOC, and (d); 25 NTU of turbidity, 8 ppm of DOC.
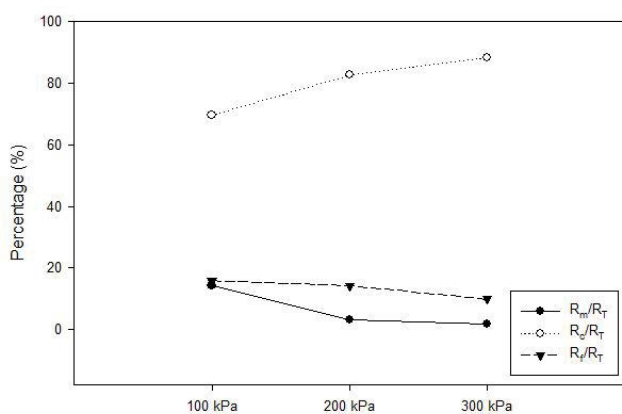

Fig. 4. Hydraulic resistance percentages according to operating pressure at case 2 (25 NTU of turbidity and 2.5 ppm of DOC) using physical backwashing.

absorption [17,18]. The average SUVA$_{254}$ values of the raw water are 9.7 L/mg-M for DOC of 2.5 ppm, and 13.9 L/mg-M for DOC of 8 ppm. It has been confirmed that the hydrophobicity of the raw water is great through the SUVA$_{254}$. In case of hydrophobic organic matter (especially with a large distribution of polymer organic matter), the adsorption rate of the organic matter for the membrane made by hydrophobic

material generates faster than that of the hydrophilic material membrane [7]. The monolithic ceramic membrane made of Al$_2$O$_3$ has a hydrophilicity because it releases the hydroxyl group (–OH) on the membrane surface. However, it was emerged that easily has occurred solute adsorption to membrane pores by the raw water having high hydrophobicity. For 2.5 ppm of DOC, J/J_0 (%) rapidly fouled membrane after V/V_0 (%) of 30%, and in case of 8 ppm of DOC, J/J_0 (%) reached 10% within 3 min at high operating pressures (200 and 300 kPa). In Jermann's study, kaolin and humic acid have been reported to exhibit a synergistic fouling effect. Membrane fouling by kaolin had a relatively large porosity of the cake layer. However, when kaolin and humic acid incorporated into the membrane, humic acid has been adsorbed on the particles to stabilize the particles, reduce the agglomerate size, and smooth the surface heterogeneity, thereby significantly reducing the porosity of the fouling layer. Thus, the associations of membrane-humic acid-kaolinite have caused irreversible membrane fouling [19]. Based on this, it was found that acute membrane fouling is formed because the compact cake layer deepens the membrane adsorption by interaction with kaolin and humic acid. In addition, monolithic ceramic membrane has been estimated to increase the efficiency of organic matter removal through the immobilization of floc by facilitating aggregation near the membrane surface by shear-induced lift force [9]. Table 4 shows the change of parameters before

Table 4
Characteristics of water before and after filtration

		2.5 ppm of DOC		8 ppm of DOC	
		Before filtration	After filtration	Before filtration	After filtration
DOC (mg/L)	Range	2.32–2.69	2.06–2.54	7.29–8.56	4.55–5.67
	Average	2.49	2.20	7.77	5.04
UV_{254} (cm^{-1})	Range	0.20–0.28	0.09–0.15	1.06–1.09	0.46–0.60
	Average	0.24	0.12	1.08	0.54
$SUVA_{254}$ (L/mg m)	Range	7.77–11.48	3.74–6.91	12.69–14.59	9.25–12.14
	Average	9.66	5.35	13.85	10.67

and after filtration. DOC had a difference in the removal efficiency according to the concentration difference, while UV_{254} all showed a removal efficiency of 50% or more. It was estimated that the UV_{254} value was lowered as the structure absorbing UV light such as a double bond in the molecule changes to a single bond by proceeding with filtration.

3.2. Comparison of irreversible membrane fouling through RISM analysis

The RISM was applied to confirm the degree of membrane fouling and the change of the resistance value according to the cleaning method. The membrane resistance, R_m, was computed by the flux measured when DI water was filtered for 1 min on the membrane initialized by CIP before the raw water was filtered. Total resistance, R_T ($R_m + R_c + R_f$), was calculated using the flux at the end of filtration and the total contamination resistance ($R_c + R_f$) was calculated by subtracting membrane resistance from total resistance. Internal fouling resistance (R_f) was calculated by subtracting R_m from the resistance value that is calculated by measuring flux using DI water after carrying the membrane cleaning out. The cake layer resistance (R_c) was calculated by subtracting the internal fouling resistance (R_f) from the total contamination resistance ($R_c + R_f$). The initial flux (J_0) with pressure were 1,491.4–1,542.9 LMH at 100 kPa, 2,468.6–2,880.0 LMH at 200 kPa, and 3,497.1–4,114.3 LMH at 300 kPa. The difference in flux at the same pressure was influenced by temperature. The Kozeny–Carman equation demonstrated that high-viscosity fluids caused low flux [20]. An increase in water temperature decreases the viscosity, which could increase the flux. It was confirmed that the initial flux has risen due to temperature changes in the experimental environment.

Fig. 4 represents the change in hydraulic resistance according to pressure as a percentage in case of physical backwashing. In case 2 (25 NTU of turbidity and 2.5 ppm of DOC), the total resistance (R_T) according to pressure was 1.60×10^{12} m^{-1}, 8.32×10^{12} m^{-1}, and 1.73×10^{13} m^{-1}, respectively. In spite of same property raw water, an increase in operating pressure caused an increase in R_T by the compressed fouling layer because an increase of transmembrane pressure contributes deposited foulants [21]. Through this, it is considered that as the operating pressure increases, the total contamination resistance ($R_c + R_f$) increases, and the ratio of membrane resistance (R_m) to gradually decreases (3.1% to

1.7%). Raw water containing kaolin-humic acid was cake resistance (R_c)-dominant. The cake resistance of the ceramic membrane was more dominant as the pressure increases, which was regarded to have a large effect of R_c on the increase of the total contamination resistance.

Fig. 5 shows the change in the membrane contamination resistance according to the cleaning method through the ratio of R_c/R_f. At 2.5 ppm of DOC, R_c/R_f increased about three times for steam cleaning and 4.3 times for CESC compared to physical backwashing. It was deliberated that irreversible membrane fouling was converted to reversible fouling through an enhanced cleaning method. Therefore, it is considered that steam cleaning and CESC can complement or substitute physical backwashing. In case of 8 ppm of DOC, R_c/R_f increased about 2.5 times for steam cleaning and 8.4 times for CESC compared to physical backwashing. CESC represented a significant increase in ratio compared to steam cleaning. It suggested that CESC was more efficient than steam cleaning in high organic matter. The RISM of case 3 (turbidity of 10 NTU, DOC of 8 ppm) is displayed in Table 5. Total contamination resistance should equal to the same pressure because they have the identical raw water property. On average, the total contamination resistance values of 9.69×10^{11} m^{-1} at 100 kPa, 8.59×10^{12} m^{-1} at 200 kPa, and 3.96×10^{13} m^{-1} at 300 kPa indicated similar membrane fouling at the same pressure. As
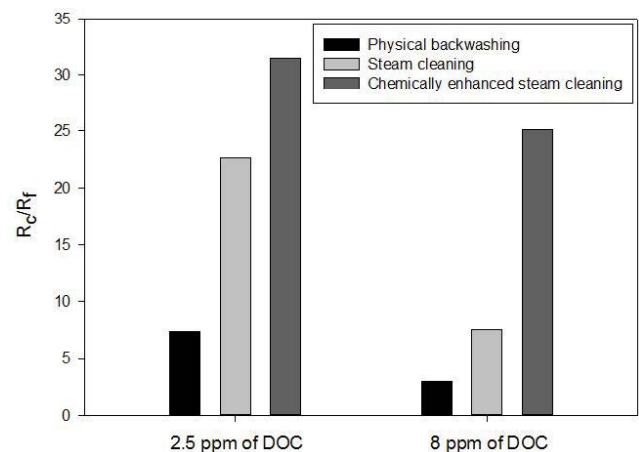

Fig. 5. Ratio of cake layer resistance (R_c) and internal fouling resistance (R_f) at operating conditions of 100 kPa, 10 NTU.

Table 5
Resistance in series model (RISM) of case 3 (turbidity of 10 NTU, DOC of 8 ppm): *B* is physical backwashing, SC is steam cleaning, and CESC is chemically enhanced steam cleaning

Operating pressure (kPa)	Cleaning method	Resistance (m⁻¹)				
		R_T	R_m	R_c	R_f	$R_c + R_f$
100 kPa	*B*	1.19 E+12 (100%)	2.26 E+11 (19.0%)	7.24 E+11 (60.7%)	2.42 E+11 (20.3%)	9.66 E+11 (81.0%)
	SC	1.14 E+12 (100%)	2.35 E+11 (20.7%)	7.95 E+11 (70.0%)	1.06 E+11 (9.3%)	9.01 E+11 (79.3%)
	CESC	1.26 E+12 (100%)	2.18 E+11 (17.2%)	1.00 E+12 (79.6%)	4.00 E+10 (3.2%)	1.04 E+12 (82.8%)
200 kPa	*B*	8.86 E+12 (100%)	2.70 E+11 (3.0%)	7.09 E+12 (80.0%)	1.50 E+12 (16.9%)	8.59 E+12 (97.0%)
	SC	9.09 E+12 (100%)	2.75 E+11 (3.0%)	8.58 E+12 (94.3%)	2.39 E+11 (2.6%)	8.82 E+12 (97.0%)
	CESC	8.64 E+12 (100%)	2.64 E+11 (3.1%)	8.22 E+12 (95.2%)	1.53 E+11 (1.8%)	8.37 E+12 (96.9%)
300 kPa	*B*	3.99 E+13 (100%)	2.91 E+11 (0.7%)	3.33 E+13 (83.3%)	6.36 E+12 (15.9%)	3.96 E+13 (99.3%)
	SC	3.99 E+13 (100%)	2.93 E+11 (0.7%)	3.92 E+13 (98.3%)	3.94 E+11 (1.0%)	3.96 E+13 (99.3%)
	CESC	3.99 E+13 (100%)	2.89 E+11 (0.7%)	3.94 E+13 (98.9%)	1.69 E+11 (0.4%)	3.96 E+13 (99.3%)

the pressure increased, the ratio of the membrane resistance decreased from 20.7% to 0.7%, and the value of total contamination resistance increased. From the data in Table 5, the resistance values variated by the cleaning methods were R_c and R_f. At each operating pressure, the value of R_c was upper in order of CESC, steam cleaning, and physical backwashing. In the steam cleaning at 300 kPa, R_c was 98.3% of the total resistance, which was 18% increased than physical backwashing. Cake layer resistance of CESC increased 18.7% compared to physical backwashing. According to the membrane fouling analysis by the resistance in series model, the cake layer which could be removed by improved cleaning seemed to be increased. Irreversible membrane fouling, which was difficult to remove by physical backwashing, was converted to reversible membrane fouling through cleaning methods including steam. Especially, R_f, which represents resistance to form contamination in the membrane, was minimized by the enhanced cleaning with NaOCl.

3.3. Recovery of fouled membrane according to cleaning methods

The membrane performance recovered via membrane cleaning by improving reduced flux. The cleaning methods were physical backwashing (*B*), steam cleaning (SC), and CESC and the results are shown in Fig. 6. The graphs were classified as 2.5 ppm of DOC and 8 ppm of DOC in order to compare the recovery rates of the membrane performance according to the difference in the quantity of filtered water. Fig. 6 shows that the cleaning efficiency tends to decrease as the operating pressure increases. Recovery rates of steam cleaning and CESC at 100 kPa of case 1 (turbidity of 10 NTU, DOC of 2.5 ppm) were 73.4% and 80.8%, respectively, that it has increased by 51.7% and 66.9% compared with the recovery rates of physical backwashing. Also, at high pressure such as 300 kPa, the steam cleaning recovery rate was 6.6 times (36.3%) of the physical backwashing recovery rate, and the CESC rate was 8.9 times (48.7%) of the physical backwashing recovery rate. It has indicated that irreversible membrane fouling, which cannot be removed by physical
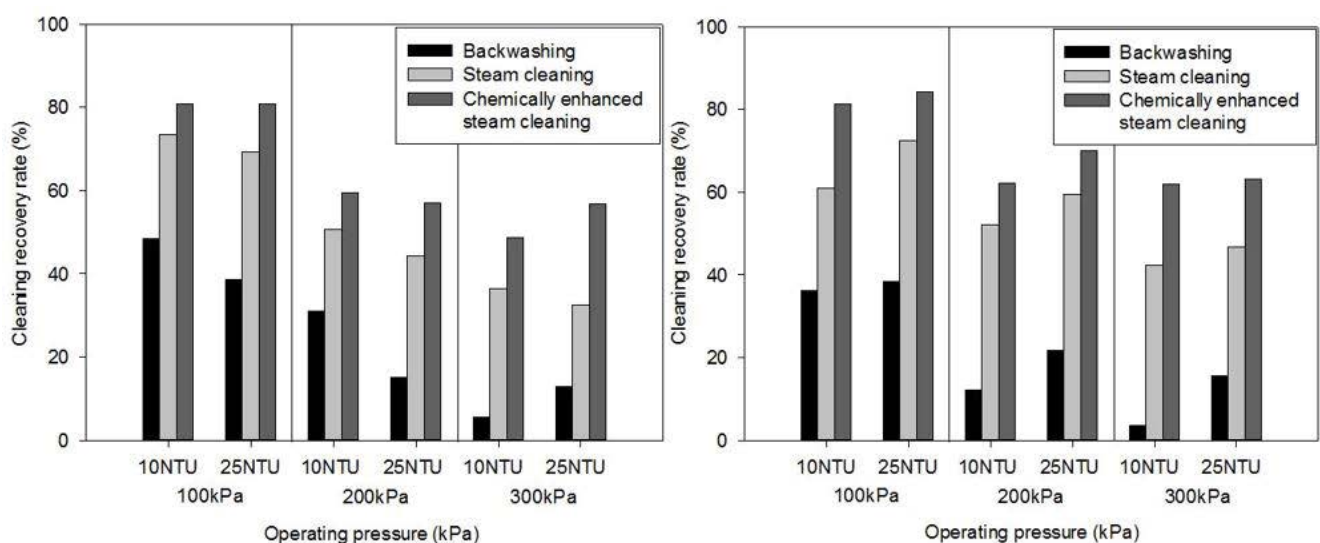

Fig. 6. Comparison of cleaning efficiency of ceramic membrane (a) is DOC of 2.5 ppm and (b) is DOC of 8 ppm.

backwashing, could be eliminated by steam cleaning and CESC.

At 100 kPa of case 3 (10 NTU of Turbidity, 8 ppm of DOC), the recovery rate of the CESC was more than 80%, which corresponds to 2.2 times of the physical backwashing. The CESC revealed a cleaning recovery rate of 60% or more at both 200 and 300 kPa. Despite the remarkably low recovery rate of physical backwashing, a high level of cleaning recovery rate in NaOCl steam cleaning indicated that NaOCl, which oxidizes organic matter, increased reaction rate between NaOCl and organic matter adsorbed in membrane pores. Wang's study has suggested that cleaning efficiency was determined by interaction active chlorine species with foulants and diffusion of active chlorine species. It has also been reported that under alkaline conditions (pH 11), OCl^- facilitated the mass transfer, while simultaneously increasing the surface negative charge and causing loosening of the matrix structure [22]. As the water temperature increases, the pH of the water decreases because the ionization constant (K_w) of water increases, so NaOCl mixed with steam at about 120°C is estimated to be washed under the influence of HOCl rather than OCl^-. Among the parameters affecting mass transfer and reaction time, the temperature

has a great influence on the recovery of membrane performance. Temperature changes the equilibrium constant of the reaction, reaction kinetics, and solubility of fouling and reactants, resulting in a structural weakening of the fouling layer [23,24]. Therefore, it was suggested that the steam at about 120°C improved the diffusion of NaOCl and increased the solubility of contaminants and detergents, representing a gradual increase in cleaning efficiency compared to steam cleaning as the cleaning time increased. As a result, it is evaluated that the rapid desorption of contaminants and the recovery rate of cleaning were increased by steam cleaning with NaOCl. At 100 kPa, the recovery rate of CESC with 10 NTU of turbidity increased 9.5% in the recovery rate of the steam cleaning despite the steam cleaning significantly eliminated contaminants. The CESC described a high level of recovery at the high-pressure operating conditions. It was 61.9% (case 3) and 63.2% (case 4) at 300 kPa. It implied an increase of 46.7% and 34.9% compared to the steam cleaning recovery rate. Thus, the cleaning methods with steam are more effective than physical backwashing by breaking the humic acid-membrane bond through pyrolysis and consequently cut down irreversible fouling. CESC can develop to upper membrane performance
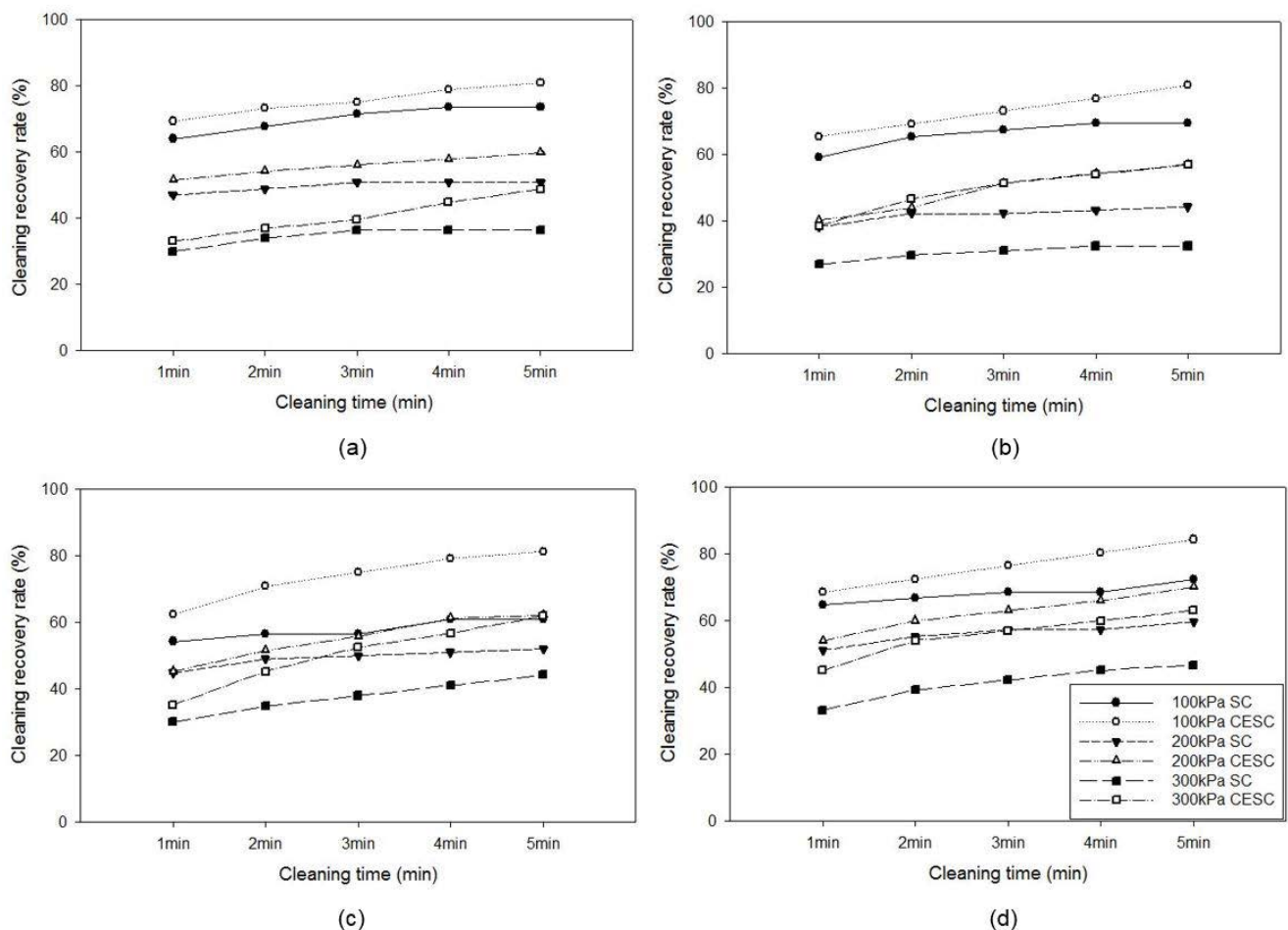

Fig. 7. Change of membrane recovery rate of steam cleaning methods with cleaning time in (a); 10 NTU of turbidity, 2.5 ppm of DOC, (b); 25 NTU of turbidity, 2.5 ppm of DOC, (c); 10 NTU of turbidity, 8 ppm of DOC, and (d); 25 NTU of turbidity, 8 ppm of DOC.

by eliminating membrane fouling caused by contaminants consolidated that are difficult to remove by steam cleaning under high pressure.

3.4. Change in efficiency with cleaning time of steam cleaning methods

In the cleaning methods with steam, the change of the membrane recovery rate with the cleaning time was observed by the steam cleaning and the NaOCl steam cleaning for 1–5 min, respectively. Fig. 7 describes a graph of the cleaning efficiency according to the cleaning time. The CESC emerged a noticeable growth of the cleaning recovery rate over time. However, it wasn't different in the cleaning time of 3 min or more in the case of the steam cleaning. It was because the dosage of NaOCl for steam cleaning increased as the cleaning time increases. In case of case 3 at 100 kPa, the steam cleaning recovery rates were 54.3%, 56.4%, 56.5%, 60.9%, and 60.9%. The cleaning recovery rate was 12.2% increase compared to 1 for 5 min. On the other hand, the recovery rate of CESC was 62.5%, 70.8%, 75.0%, 79.2%, and 81.3%, respectively. This implied 30.1% increase compared to 1 min when cleaning for 5 min. The CESC can break the limit of steam cleaning by recording a higher recovery rate than the steam cleaning at the same cleaning time, and increasing the recovery rate as the cleaning time becomes longer. When analyzed by a scatter plot, the slope of the trend line for the CESC was higher than that of the steam cleaning, and the R^2 value of the trend line for CESC had a value closer to 1, which indicates a relatively linear graph. According to the slope of the trend line, about 16.3 min at steam cleaning was required to obtain the same cleaning efficiency as CESC for 5 min at 100 kPa of case 3. It required about 3.3 times of cleaning time compared to the CESC. However, as described above, it is expected that the steam cleaning will require longer than about 16.3 min since it does not greatly affect the recovery rate after a certain time. The recovery rate of the CESC tends to be soared at 300 kPa. NaOCl, which oxidizes the organic matter, was contacted with the steam in the ceramic membrane, effectively detaching the organics that had been consolidated. Therefore, it is considered that the CESC will be able to effectively cope with the severe membrane fouling generated suddenly under extreme operating conditions such as a high concentration of organic matter and high pressure by adding a little concentration of chemicals.

4. Conclusions

In this study, the cleaning efficiency of CESC, which combined with NaOCl and steam, was compared with other cleaning methods according to diverse raw water conditions. Under high pressures of 200 or 300 kPa, the rate of flux reduction soars after the middle stage of filtration. Synergistic effects of particles and organic matter in artificial raw water resulted in compact adsorption and acute irreversible fouling under extreme operating conditions. In terms of RISM, the resistance was R_c is the dominant for raw water incorporated with kaolin and humic acid, then the effect was more dominant as operating pressure increased. The cleaning methods including steam indicated

a remarkable cleaning recovery rate as compared with other cleaning methods. CESC could minimize the amount of chemicals used in membrane cleaning as well as increase the cycle of CIP by using a concentration equivalent to 1/300 of the NaOCl concentration applied in CIP. CESC requires more energy and establishment cost than physical backwashing due to a steam generator. However, CESC allows for a flexible response to rapidly originating membrane fouling and enables fast recovery of membrane performance even with short cleaning time. Therefore, it has the advantage of reducing the time and chemicals cost of cleaning and is expected to help improve the operating efficiency of water purification plant if the concentration of NaOCl used in cleaning and the cleaning time are appropriately adapted according to the degree of membrane fouling.

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