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Steam cleaning for control of membrane fouling in ceramic membrane system

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A B S T R A C T

In this study, the recovery efficient by operating pressure and influent water quality was evaluated in order to develop cleaning process for ceramic membrane using high temperature steam. The operating pressure was fixed at 100, 200, and 300 kPa during filtration time and the change in permeate flux and resistance was measured over time. The artificial water was made by adjusting the concentration of turbidity (25, 50, 100 NTU) and dissolved organic carbon (DOC) (4, 8 mg/L) using kaolin and humic acid. As a result of evaluating the steam injection time for 0–5 min in order to derive the optimum conditions of steam cleaning, the efficiency of 35%–54% was improved compared with the physical washing when steam was injected for 3 min. It was confirmed that steam cleaning at high turbidity and high organic matter condition increased cleaning recovery rate of 15.1%–53.8% compared with physical backwashing. As a result of analyzing the membrane fouling formed by using the resistance in series model, it was confirmed that the steam cleaning improves the cleaning efficiency by converting 3.8%–39.2% of irreversible fouling (R_f) into reversible fouling (R_c) depending on conditions. In the ceramic membrane process, the steam cleaning showed better efficiency than the physical backwashing. Steam cleaning can be expected to improve the stability of the process and reduce the chemical costs.

Keywords: Ceramic membrane; Steam cleaning; Membrane fouling; Membrane resistance; Water treatment

1. Introduction

The introduction of MF (microfiltration) and UF (ultrafiltration) in the water treatment process makes it easier to obtain stable water quality and quantity than conventional water treatment processes. Also it has the advantage of being able to effectively remove pathogenic microorganisms such as Cryptosporidium and Giardia, thereby producing high quality drinking water [1–3]. Currently, water treatment processes using membrane filtration mainly use organic membranes, but it is difficult to operate at high temperature and high pressure, and biological and chemical stability are needed. Therefore, it is an increasing demand for the development of ceramic membranes that can be operated stably for a long time on severe conditions [4–6]. Ceramic membrane can maintain the characteristics under high temperature and high pressure and has a simpler shape and configuration than the organic membrane. It is easy to clean when it is contaminated, so it can maintain high concentration and high transmittance constantly, and it is applicable to high turbidity and high viscosity solution. It has excellent chemical resistance and durability, so that even if exposed to organic solvent or acid-alkali solution for a long time, the membrane is less damaged. In addition, since the pH range is stable from 1 to 14, it can be applied to strong acid and base solutions for chemical cleaning [7–9]. The MF/UF membrane filtration process is the main mechanism of membrane filtration due to the porosity of the membrane and the size of the contaminants. Particle matter and dissolved organic substances separated by pores or deposited on the surface of the membrane cause a reduction

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in the quantity of water treated in the membrane filtration process and a decrease in operating efficiency such as an increase in transmembrane pressure (TMP) [10,11]. The membrane whose performance is deteriorated by the fouling in the filtration process is restored by physical or chemical cleaning depending on the degree of fouling. Physical backwashing is a method of recovering the performance of the membrane by removing fouling matter adsorbed to the membrane surface by using air, filtration water. In general, physical backwashing is included in the filtration process, it can prevent the compaction of the membrane fouling layer and reduce membrane fouling, thereby improving the permeation flux [12]. Chemical cleaning removes membrane fouling on organic and inorganic materials using acid and base chemicals to remove contaminants accumulated on the membrane that cannot be removed by physical backwashing. Chemical cleaning is classified as chemically enhanced backwashing (CEB) and cleaning in place (CIP), and CEB was performed in a short period of time using a low chemical concentration compared with CIP. The CEB process is applied when the water quality of the raw water changes temporarily or as the TMP increased. CIP is applied when the CEB efficiency is to be reduced or the membrane is to be restored to its initial state. The CIP process requires a longer time to clean than the CEB, frequent chemical cleaning reduces the operation rate of the membrane facility. In addition, corrosion of the pipe may be caused, and the characteristics of the membrane may be modified to cause a problem of performance deterioration. Membrane fouling control is very important in the membrane filtration process because the cost of chemicals required for chemical cleaning increases the total operation cost and production cost [13,14].

In this study, the cleaning technology using high temperature steam was developed and evaluated for applicability to ceramic membranes. The saturated steam of high temperature facilitates pyrolysis of the oil more easily, and the temperature of the saturated steam becomes higher as the pressure increases. Thermal cracking occurs in which the binding force of contaminants adsorbed on the surface of the membrane due to the steam is weakened. Thermal cracking is a phenomenon in which the combined forces between molecules weaken as the temperature increases. When the entropy (degree of freedom) increases more than the enthalpy (internal energy) by the endothermic reaction, that is, when the degree of freedom of the molecule becomes larger, it becomes more active. As the temperature increases, the density of the vapor increases and the enthalpy required to convert the liquid to gas increases. The more negative the free energy, the more active the endothermic reaction and the better the thermal cracking. Since the steam cleaning can be applied at a temperature of more than 100°C, it was evaluated by applying it to a ceramic membrane having an advantage of thermal stability. Ceramic membranes are known to have durability against temperatures above 1,500°C. In this study, the applicability of steam cleaning was evaluated by comparing the efficiency of steam cleaning and physical backwashing. In addition, the recovery rate according to the operating conditions and raw water quality was evaluated and the optimum steam cleaning conditions were derived.

2. Materials and methods

2.1. Water quality of artificial water

In this study, artificial water was used to confirm the effect of turbidity and organic matter on the formation of membrane fouling. Kaolin (Showa, Japan) and humic acid (Fluka, Germany) were added to adjust the feed water for turbidity (10, 25 NTU) and organic matter (2.5, 10 mg/L). The humic acid stock solution was made by dissolving humic acid in deionized water dissolved by heating at 80°C for 30 min for easy dissolution of humic acid. Then, the solution was filtered in polyphenylsulfone magnetic filter funnel (Pall, USA) using a microfiber filter (Whatman, USA) with a mean pore size of 0.45 μm. The DOC of the stock solution was analyzed by TOC analyzer (TOC-V, Shimadzu, Japan).

2.2. Experimental set-up

The monolith type ceramic membrane used in the experiment has a pore size of 0.1 μm and the characteristics of the ceramic separator are shown in Table 1. The operation pressure was fixed at 100, 200, and 300 kPa and operated by the constant pressure filtration method in order to check the membrane fouling difference according to the operating pressure condition. The ceramic MF system used in the batch experiment is illustrated in Fig. 1, and the change in flux with time was measured during filtration of 15 L of raw water. Flux changes were continuously measured by measuring the mass of the filtered water using electronic balance. The steam generator is designed to check the recovery of membrane contamination by steam cleaning. It can generate steam by heating the water to maintain the set temperature.

The detailed conditions of the filtration and washing process are shown in Table 2. In the filtration process, 15 L of raw water was filtered on each pressure condition, and the recovery rate of the fouled membrane was confirmed by physical backwashing and steam cleaning. Physical backwashing used water and air at pressure of 500 kPa to remove the fouling material accumulated on the membrane surface. Steam cleaning was performed by injecting steam of 120°C for 1–3 min to compare the recovery rates according to the steam injection time. The specific volume of 120°C steam is 0.8857 m^3/kg, the injection pressure is 100 kPa and the steam flow rate is about 15 m/h.

2.3. Calculation of membrane fouling using resistance-in-series (RIS) model

In order to quantitatively analyze the membrane fouling, the water permeability through the application of filtration resistance model (resistance in series) from Darcy's law is shown as the respective resistances. Fouling resistance is divided into intrinsic resistance of membrane (R_m), reversible fouling resistance which forms fouling on membrane surface (R_c, cake layer resistance), and irreversible fouling resistance which forms fouling inside membrane pore (R_f, internal fouling resistance). The schematic diagram for each resistance is shown in Fig. 2. Flux (J) and TMP of each resistance value have the relations as Eqs. (1)–(3). The flux is proportional to the driving force TMP on the membrane,

Table 1
Characteristics of ceramic membrane

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 channel	55	
Membrane surface area	0.035 m^2	
pH range	1–14	
Maximum operating pressure	20 kgf/cm^2	
Filtration type	Dead-end	
Manufactory	Metawater, Japan	

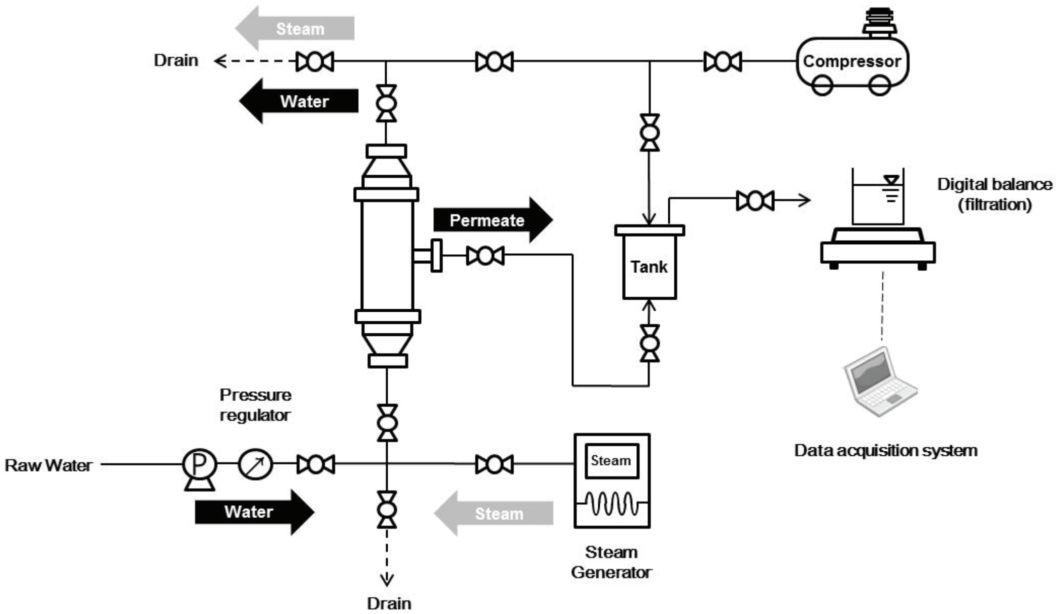

Fig. 1. Schematic diagram of lab-scale ceramic MF system.

and is inversely proportional to the product of the total resistance (R_T) and viscosity (μ) of the treated water. The membrane fouling analysis using the series filtration resistance model is shown in Fig. 3, the intrinsic membrane resistance (R_m) of the membrane was measured using distilled water for 10 min before the filtration. The flux change during filtration of the raw water was measured and the resistance (R_c) due to the reversible fouling recovered through cleaning was measured. The resistance (R_f) due to irreversible fouling, which was not recovered by cleaning, was measured as comparing the intrinsic resistance of the initial membrane by filtering again with distilled water for 10 min after cleaning.

$$J = \frac{\text{Driving force}}{\sum \text{Resistance}} = \frac{\text{TMP}}{\mu \times R_T} \quad (1)$$

$$R_T = R_m + R_c + R_f \quad (2)$$

$$\text{J} = \frac{\text{TMP}}{\mu\left(R_m + R_c + R_f\right)} \quad (3)$$

Table 2
Specific conditions of operating and cleaning process

Process	Time	Mode	Specific conditions
Filtration	60–120 mins	Running time	– Filtration of 15 L
			– Dependent on feed conditions and operating pressure
Steam cleaning	0 – 3 mins	Vapor temperature	120 °C
		Injection pressure	100 kPa
		Injection velocity	15 m/h
Physical backwashing	1 min	Standby	10 s
		Pressurization	20 s
		Water backwash	500 kPa, 3 s
		Air blow	500 kPa, 1s
		Pressure relief	5 s
		Stop	10 s
		Valve on/off time	11 s

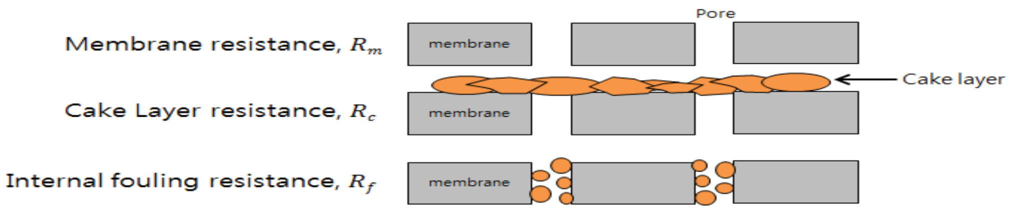

Fig. 2. Diagram of reversible and irreversible fouling formation [15].

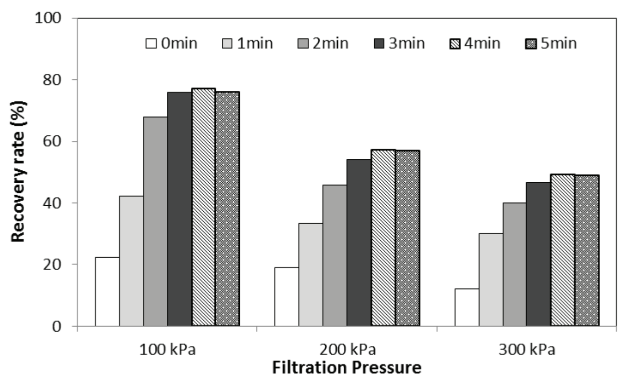

Fig. 3. Comparison of cleaning efficiency by steam injection time.

3. Results and discussion

3.1. Effect of steam injection time for cleaning membrane

In order to evaluate the recovery of the fouled ceramic membrane using steam, steam was injected into the contaminated membrane for 0–5 min to confirm the efficiency of steam cleaning by injection time. The water quality of the raw water was 25 NTU and DOC 2.5 mg/L, and it was filtered at the pressures of 100, 200, and 300 kPa to compare the cleaning efficiency according to the operating pressure. To compare the efficiency of the physical backwashing process and the steam cleaning, an evaluation was performed according to physical backwashing (0 min) and steam injection time (1–5 min). After the end of the experiment, CIP was performed to restore the membrane state to the initial state.

After the filtration process, physical washing and steam cleaning results are shown in Fig. 3. Regardless of the operating pressure, 0 min of physical backwashing showed a recovery rate of less than 20%, but when steam was injected for 1 min, additional recovery of about 15%–20% was confirmed. As a result of injecting steam for 3 min, it was recovered by about 47%–76% according to the operating pressure, and it was confirmed that the efficiency of about 35%–54% was improved compared with physical backwashing. As a result of injecting steam for 5 min, it was confirmed that it was about 1%–3% higher than the result of 3 min, and the optimum injection time was determined to be 3 min.

3.2. Comparison of flux decrease rate by turbidity and DOC concentrations

To verify the efficiency of steam cleaning for turbidity and DOC concentration conditions, the experiment was repeated by adjusting the turbidity and DOC concentration of the raw water. Turbidity concentrations were adjusted to 10 and 25 NTU, and DOC concentrations were adjusted to 2.5 and 10 mg/L, respectively, to compare steam cleaning efficiencies on each condition. The flux decrease rate during the filtration of 15 L of raw water is shown in Figs. 4–6. Fig. 4 shows the results of filtration of raw water of 10 NTU turbidity and 2.5 mg/L DOC at operating pressures of 100, 200, and 300 kPa. During the filtration of 15 L, the flux decreased by 66.7%–87.2% depending on the operating pressure. The turbidity concentration was adjusted to 25 NTU (DOC 2.5 mg/L) to evaluate the effect of turbidity concentration on the flux change (Fig. 5). When the turbidity concentration increased, the flux reduction rate by operating pressure was 81.1%–87.9%, which was decreased by 0.7%–14.4% compared with 10 NTU. It was also confirmed that the flux decreased rapidly when the turbidity concentration increased. When the turbidity is reduced flux by 50% at 10 NTU, the volume rate is 75%, 55%, and 50%, depending on the operating pressure. When the flux was reduced by 50% at turbidity 10 NTU, the volume rate was found to be 75%, 55%, 50%, while for turbidity 25 NTU, the volume rate was verified to be 20%–30%. DOC concentration was adjusted to 10 mg/L (turbidity 10 NTU) in order to confirm the effect of DOC concentration on flux change (Fig. 6). The flux reduction rate was 87.7%–95.2%, which was 21.0%–8.0% lower than DOC concentration 2.5 mg/L (turbidity 10 NTU).

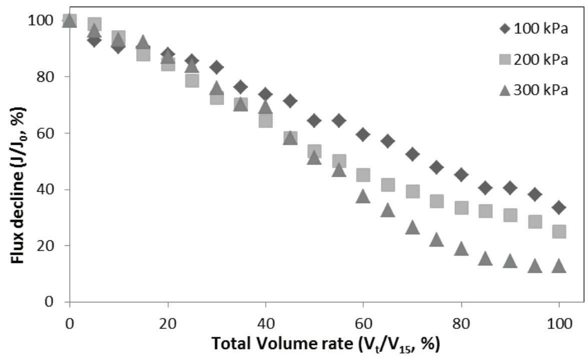

Fig. 4. Flux decrease rate at turbidity 10 NTU by operating pressure.

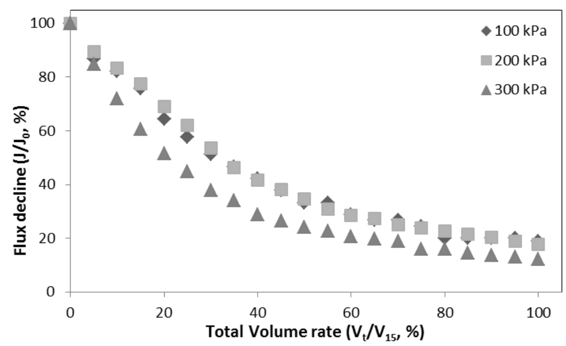

Fig. 5. Flux decrease rate at turbidity 25 NTU by operating pressure.

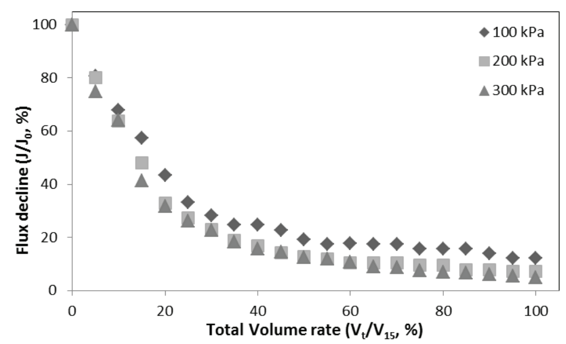

Fig. 6. Flux decrease rate at DOC 10 mg/L by operating pressure.

The flux reduction rate was 50% to 15% to 20% of the volume ratio, and the flux decreased about 35%–55% faster than the DOC 2.5 mg/L (turbidity 10 NTU). As a result, it was confirmed that the flux decrease was increased as the operating pressure increased and the flux decreased at the beginning of operation. Also, as the turbidity and DOC concentration increases, the initial flux reduction rate is further increased.

3.3. Recovery efficiency of steam cleaning and physical backwashing

Physical backwashing and steam cleaning were performed on the fouled membrane by raw water quality and operating pressure condition. The recovery rate evaluation results are shown in Figs. 7–9. Physical backwashing was carried out with water and air at pressure of 500 kPa. Steam cleaning was carried out with steam at 120°C for 1–3 min. As a result of the experiment using raw water of 10 NTU (DOC 2.5 mg/L) of turbidity, the recovery rate by physical backwashing was 32.4%–67.9%, and the physical backwashing recovery rate decreased with increasing operating pressure. When steam washing was performed for 3 min, recovery rate of about 11.7%–19% was increased compared with physical backwashing. The turbidity was adjusted to 25 NTU (DOC 2.5 mg/L) and the results are shown in Fig. 8. When the turbidity concentration increased, the physical washing recovery rate decreased from 20.3% to

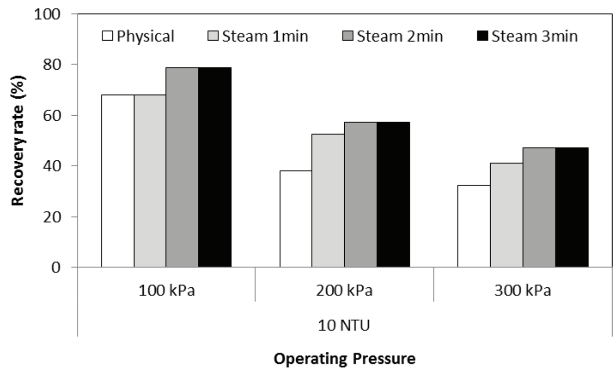

Fig. 7. Comparison of cleaning efficiency by operating pressure (turbidity 10 NTU, DOC 2.5 mg/L).

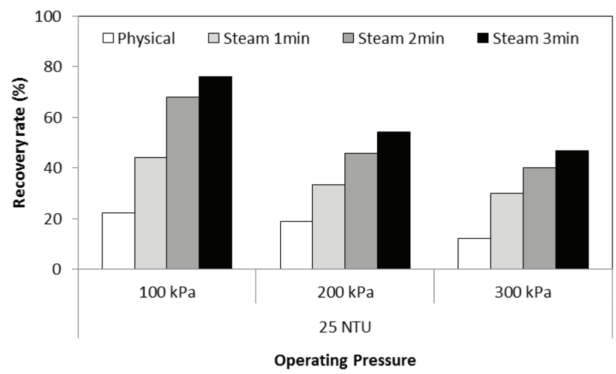

Fig. 8. Comparison of cleaning efficiency by operating pressure (turbidity 25 NTU, DOC 2.5 mg/L).

45.7%, from 12.1% to 22.2%, but the recovery rate increased to 46.7%–76.0% when steam was injected for 3 min. As a result of evaluating the DOC concentration at 10 mg/L (10 NTU of turbidity), the physical backwashing recovery rate decreased to 18.8%–44.7%, but it increased to 43.6%–64.8% in the steam cleaning. The particulate and organic matter contained in the raw water are adsorbed on the membrane surface or pores, reducing the physical backwash efficiency. However, when the steam is injected, it is considered that the efficiency of the steam cleaning is improved by weakening the bonding of the foulants due to thermal cracking.

3.4. Analysis of membrane fouling formation using resistance in series model

The membrane fouling formed applying the experimental results by operating condition and water quality condition to resistance in series model were classified into reversible (R_c) and irreversible (R_f) fouling. The results of physical backwashing and steam cleaning (3 min) are shown in Figs. 10–12. The ratio of reversible fouling (R_c) to total resistance (R_T) in the results of turbidity 10 NTU (DOC 2.5 mg/L) is 53.3%–68.8%.

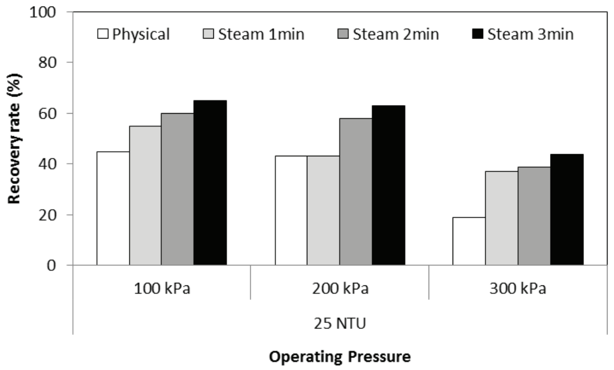

Fig. 9. Comparison of cleaning efficiency by operating pressure (turbidity 10 NTU, DOC 10 mg/L).

When steam cleaning was applied, about 3.8%–9.9% of irreversible fouling (R_f) was converted to reversible fouling (R_c) (Fig. 10). In the results of the turbidity 25 NTU (DOC 2.5 mg/L) shown in Fig. 11, the ratio of irreversible fouling (R_f) was 35.0%–41.2%. However, when steam cleaning was applied, about 23.3%–39.2% of irreversible fouling (R_f) was converted to reversible fouling (R_c). As a result of steam cleaning, the ratio of reversible contamination (R_c) among total resistance (R_T) was 70.0%–79.2%. At the DOC 10 mg/L (turbidity of 10 NTU), the highest flux reduction rate (92%) was obtained during the filtration process, which increased the total resistance (R_T). The reversible contamination rate (R_c) of the total resistance was the highest at 87.2% (Fig. 12). When the turbidity concentration increased, the irreversible contamination (R_f) resistance increased about 2–6 times from $1.32 - 7.47 \times 10^{11}$ m^{-1} to $0.79 - 1.57 \times 10^{12}$ m^{-1}. However, when steam cleaning was applied, it was found to convert about 39%–86% of the irreversible fouling (R_f) into reversible fouling (R_c). In addition, when the concentration of organic matter increased, irreversible contamination (R_f) was found to be about $0.32–1.23 \times 10^{12}$ m^{-1}, which was increased by about 1–2.4 times. In the steam cleaning, about 60% irreversible fouling (R_f) was converted to reversible fouling (R_c). It can be concluded that steam cleaning with membrane filtration using high turbidity and high organic matter water can control irreversible fouling (R_f) of about 39%–86% and enable more efficient maintenance.

4. Conclusions

The purpose of this study is to improve the cleaning efficiency by applying steam cleaning in the ceramic membrane filtration process. In order to compare the efficiency of physical backwashing and steam cleaning, membrane fouling occurred by constant pressure operation at operating pressures of 100, 200, and 300 kPa. Turbidity (10, 25 NTU) and DOC (2.5, 10 mg/L) concentrations were adjusted to confirm the efficiency of steam cleaning according to water quality conditions. The conclusive remarks are as follows:

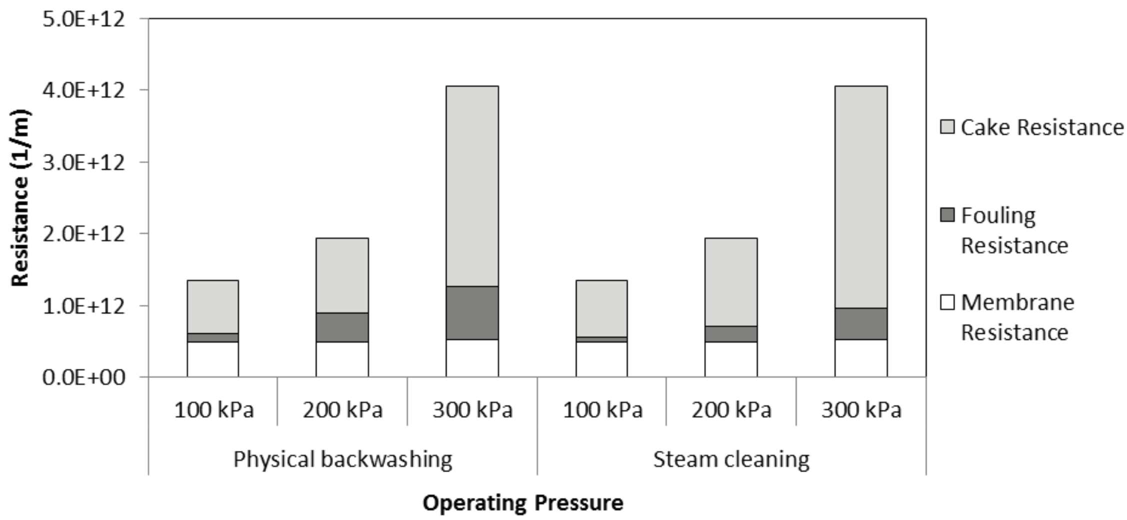

Fig. 10. Comparison of membrane fouling formation by physical backwashing and steam cleaning (turbidity 10 NTU, DOC 2.5 mg/L).

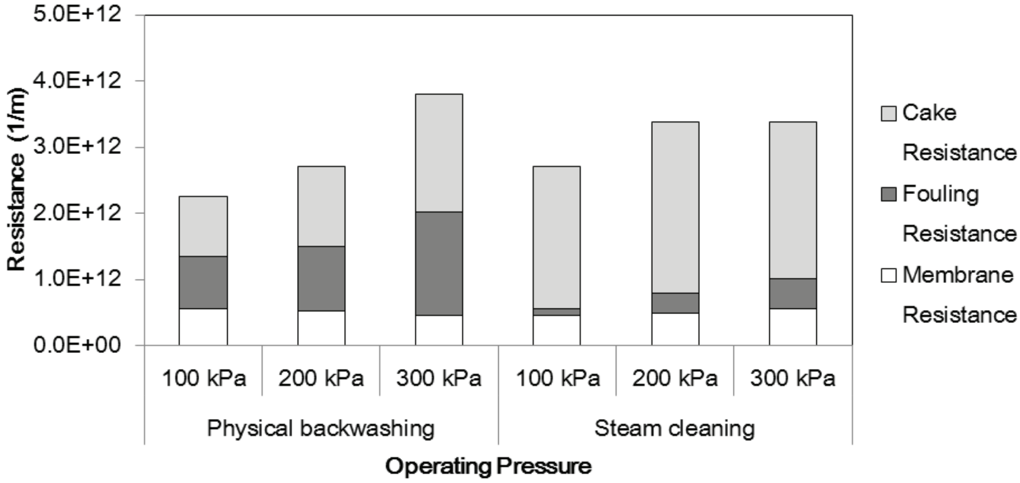

Fig. 11. Comparison of membrane fouling formation by physical backwashing and steam cleaning (turbidity 25 NTU, DOC 2.5 mg/L).

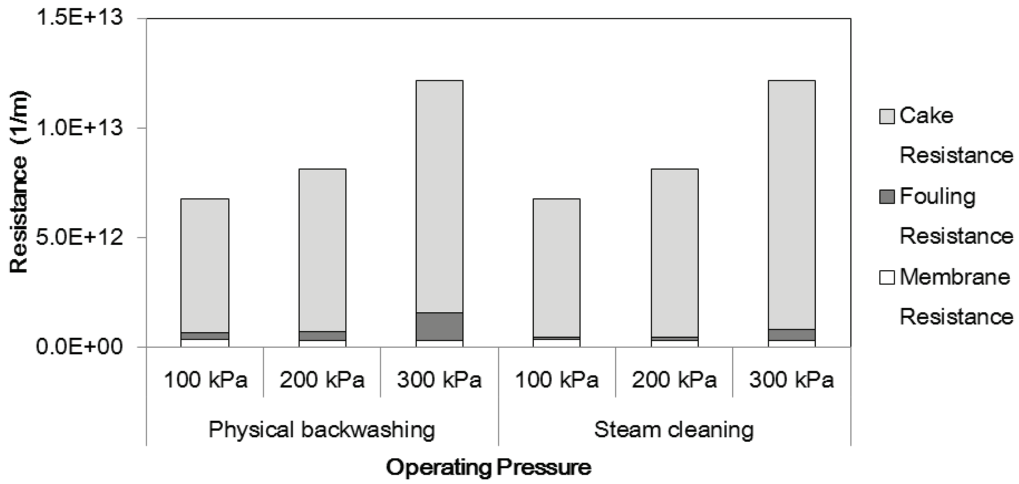

Fig. 12. Comparison of membrane fouling formation by physical backwashing and steam cleaning (turbidity 10 NTU, DOC 10 mg/L).

1. As a result of evaluating the steam injection time for 0–5 min in order to derive the optimum conditions of steam cleaning, the efficiency of 35%–54% was improved compared with the physical washing when steam was injected for 3 min.
   After 3 min, it increased by 1%–3%, but the effect was not significant, so the optimal injection time was determined to be 3 min.
2. As a result of the high turbidity filtration test, the recovery by physical backwashing was less than 22%, and the efficiency of 19.1%–45.7% was improved when steam cleaning was applied for 3 min. Steam cleaning showed 10.7%–43.8% higher recovery rate than physical backwashing depending on water quality and operating conditions. Also, it was confirmed that the steam cleaning showed a recovery rate 15.1%–30% higher than the physical backwashing in the result of evaluation on adjusting the DOC concentration to 10 mg/L.
3. As a result of analyzing the membrane fouling formed by using the resistance in series model, it was confirmed that the steam cleaning improves the cleaning efficiency by converting 3.8% to 39.2% of irreversible fouling (R_f) into reversible fouling (R_c) depending on conditions.
4. In the ceramic membrane process, the steam cleaning showed better efficiency than the physical backwashing. Thus steam cleaning can be expected to improve the stability of the process and reduce the chemical costs.

Acknowledgment

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Symbols

J	—	Water flux, m/s
R_m	—	Membrane resistance, 1/m
R_c	—	Cake layer resistance, 1/m
R_f	—	Internal fouling resistance, 1/m
μ	—	Viscosity of permeate, kg/m/s, at 20°C water
ΔP_T	—	Trans-membrane pressure, kg/m/s^2

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