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Influence of coagulation on UF performance in a pilot-scale seawater desalination plant

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ABSTRACT

Seawater desalination has been driven by an increase in water resource scarcity and freshwater demand. Reverse osmosis (RO) is recently a popular technology for seawater desalination but suffers from problems associated with membrane fouling. Accordingly, it is crucial to apply proper pretreatment techniques to mitigate RO fouling. In this study, ultrafiltration (UF) combined with coagulation was applied to control RO fouling in a pilot-scale seawater desalination plant. Critical flux for the UF was determined without and with coagulation. The effect of feed water quality on UF performance was examined. Cleaning efficiencies for the UF membranes were compared under various conditions to explore the optimum condition.

Keywords: Reverse osmosis; Fouling; Pretreatment; Ultrafiltration; Coagulation; Critical flux

1. Introduction

Reverse osmosis (RO) is a membrane technology for the desalination of seawater, which provides a solution to the problems associated with water scarcity [1]. Compared with thermal desalination technologies such as multistage flash and multiple-effect distillation, RO uses less electrical energy, which makes it attractive as a sustainable option for desalination [2]. However, RO membranes may be crippled by foulant materials such as colloidal suspensions, particulates, dissolved organics, inorganic matter, and biofilm development [3,4]. Algal outbreaks due to climate change and seawater contamination also affect RO desalination by causing serious membrane fouling [5]. In this context, a variety of technologies have been attempted to predict [6–8], control [3,4], and retard RO membrane fouling [9,10]. One of such techniques is the cleaning of fouled RO membranes [11] and another one is the pretreatment prior to RO process [12]. Although cleaning is essential in most cases, the pretreatment is highly desired as a preventive measure against RO fouling [3,12–14]. As a result, great importance for successful operation of RO desalination plants is the application of efficient pretreatment to reduce the amount of potential foulants from the RO feed water [13,15].

Ultrafiltration (UF) is one of the most efficient techniques for the pretreatment because it can satisfy increased requirements of feed water quality for RO process [13,14,16]. Since most particles are removed by UF, it can be used without cartridge filters prior to RO system and the water treated by UF can be directly supplied to RO systems [13]. Other advantages of UF include decrease in chemical consumptions, the capability of relatively low pressure operations, removal of small microorganisms, and the stability of product water quality [17,18]. UF can be used without or with chemicals such as

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coagulants although its performance may be improved by the use of coagulants [16,18,19].

Although a handful of studies have reported UF pretreatment in laboratory-scale systems [16,19–21], relatively few works have focused on the optimization of the UF operating condition in pilot plants, which is essential to provide useful information in full-scale applications. Accordingly, the aim of this research was to explore the optimum pretreatment conditions for RO desalination system using UF in pilot plants. The influence of coagulation on specific cake resistance of the flocs and UF membrane filterability was investigated. The effectiveness of cleaning in place (CIP) for the UF membrane was examined under different conditions.

2. Experimental methods and conditions

2.1. Feed water

A pilot-scale plant was installed at GS EPS pumping station and operated using the seawater taken from the west sea in Korea. The raw seawater was fed into the cartridge filter. The temperature ranged from 6.5°C to 26.1°C and the total dissolved solids (TDS) of the seawater was in the range of 30 and 34 g/L. The raw water qualities specified in Table 1 were the average values of all experiments.

2.2. Membrane

An inside-out hollow fiber UF (Dizzer 5000, Inge, Germany) was used as the pretreatment for seawater RO process. According to the membrane manufacturer, the material of membrane was polyester with styrene and methyl methacrylate (PESM). PESM-based membrane has outstanding oxidative, thermal, and hydrolytic stability. The specific characteristics of membrane are listed in Table 2. The range of operating pressure of UF membrane system is 0–2.5 bar.

Table 1
Average raw water qualities for the tested seawater

pH	6.92–7.98
Temperature (°C)	6.5–26.1
SDI	5.11
TDS (g/L)	30–34
Turbidity (NTU)	5.2–101
Na (mg/L)	9,082–9,500

2.3. Experimental methods

Fig. 1 shows the schematic diagram of the pilot plant. The UF membrane system which has a maximum production capacity of 150 m^3/d was used. The flux was 60 L/m^2 h with the feed flow rate of 17.5 m^3/h. Under this condition, the ratio of permeate flow to feed ratio was 0.169. The backwash interval and the reverse flow interval were 30 min and 30 s, respectively. The operating conditions for UF are summarized in Table 3.

The performances of UF membrane was compared with and without coagulant. $FeCl_3$ (Iron(III)) was selected as the coagulant. Reagent grade chemicals were used for the experiments. Caustic soda and hydrogen chloride were used for the pH adjustment. Coagulation conditions determined

Table 2
Characteristics of UF membrane

Parameters	Values
Length (mm)	1,680
Material	PESM
Pore size (μm)	0.02
Active membrane area (m^2)	50
Hollow fiber diameter inner (mm)	0.9
Hollow fiber diameter outer (mm)	4.3
Capillaries per fiber	7
Maximum tolerance temperature (°C)	40
Maximum tolerance pH	13
Maximum transmembrane pressure (TMP)	2.5

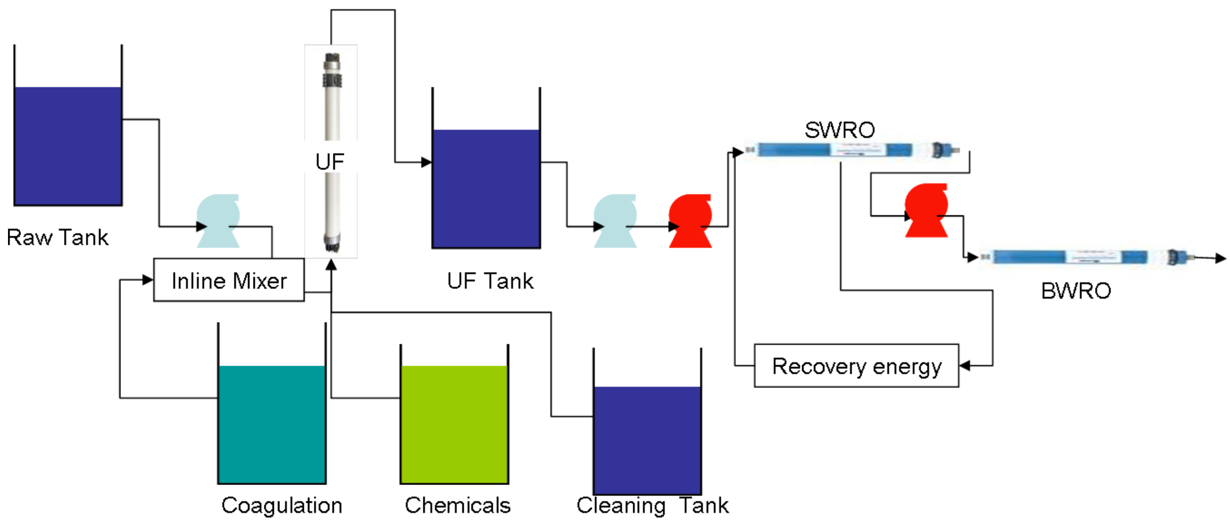

Fig. 1. Schematic diagram of the pilot plant.

Table 3
Operation condition of UF membrane

Parameters	Values
Flux (LMH, L/m²/h)	60
Feed flow rate (m³/h)	17.5
Reverse filtration flow rate (m³/h)	22
Operation mode and duration (min)	Filtration (30) → reverse filtration (0.5)

by jar-test experiments. It was applied efficient to performance inline mixer. Turbidity was measured by a turbidimeter (Hach, USA) and the silt density index-15 (SDI_{15}) was analyzed by Osmonics auto SDI tester.

3. Results and discussion

3.1. Optimum coagulant dose

Prior to UF experiments, a set of jar tests were carried out to determine the optimum $FeCl_3$ dose. As shown in Fig. 2 and Table 4, the turbidity decreases with an increase in the $FeCl_3$ dose. The turbidity ranged from 0.5 to 6 NTU with the coagulant dose range of 0–25 mg/L as Fe. Based on these results, the optimum $FeCl_3$ dose is 5 mg/L. No significant reduction in turbidity of treated water was observed above the $FeCl_3$ dose of 5 mg/L.

3.2. Effect of coagulation on UF flux decline

The changes in UF flux with time were compared between the case without coagulation and the case with $FeCl_3$ dose of 5 mg/L. The results are shown in Figs. 3 and 4. During the 500 min operation, the UF operation without coagulation resulted in a slight reduction (~25%) in reduced flux (J/J_0), indicating that UF fouling occurred. On the other hand, the UF operation with the coagulation exhibited no apparent flux decline. As expected, the coagulation helps to increase the stability of UF operation for seawater pretreatment. A possible reason for this is the reduction in particle concentrations by the coagulation as shown in Fig. 2. Another possibility is the change in the properties of cake formed from suspended particles in the feed water [18,22].

3.3. Specific cake resistances

The specific cake resistances of suspended particles in the feed waters were estimated using the cake filtration equation [22]:

$$\frac{t}{V} = \frac{\alpha \eta c}{2A^2 \Delta P} V + \frac{R_m \eta}{A \Delta P} \quad (1)$$

where t is the test time, V is the volume of the permeate, α is the specific cake resistance, c is the dry mass of filter cake per permeate volume, ΔP is the transmembrane pressure (TMP), h is the permeate viscosity, and A is the membrane area. The experimental results are shown in Fig. 5 and the specific cake resistances are summarized in Table 5. With the addition of the coagulant, the specific cake resistance decreases and shows its minimum with the dosage of 5 mg/L.

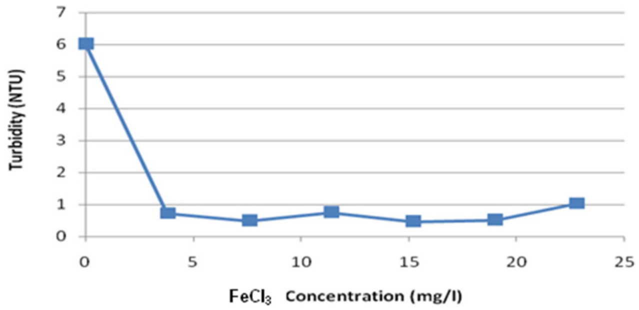

Fig. 2. Effect of coagulant dose on turbidity of the treated water in jar tests.

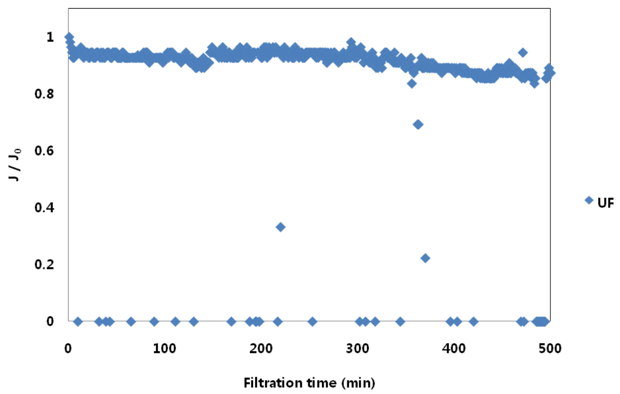

Fig. 3. Dependence of reduced flux (J/J_0) on filtration time for feed water without coagulation.

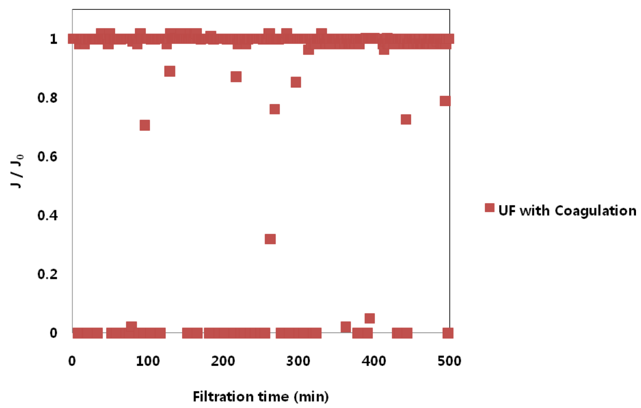

Fig. 4. Dependence of reduced flux (J/J_0) on filtration time for feed water with coagulation ($FeCl_3$ dose of 5 mg/L).

According to the Kozeny–Carman equation [23,24], the specific cake resistance (α) for a spherical particle is given by:

$$\alpha = \frac{180(1-\varepsilon)}{\varepsilon^3 d^2} \quad (2)$$

where ε is the cake porosity and d is the diameter of the particle. By adding coagulant, the size of the particles in the feed water increases, leading to a decrease in the specific cake resistance. However, not only the particle size but also the porosity affects the specific cake resistance and the specific

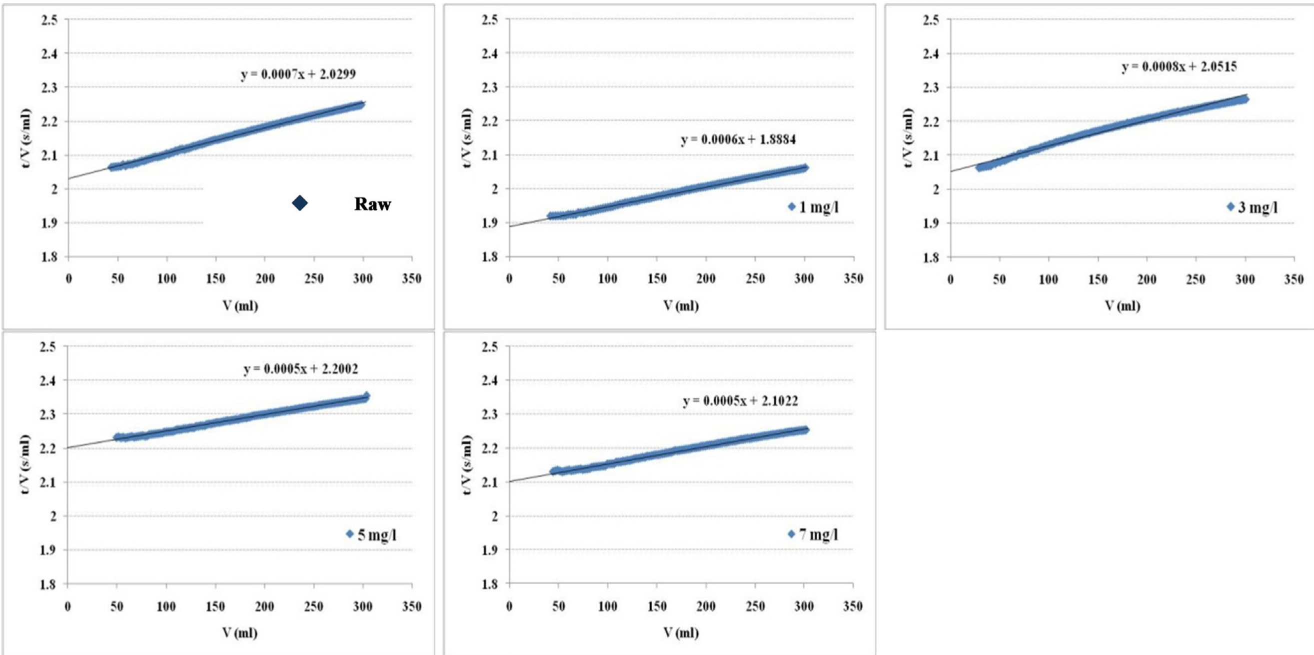

Fig. 5. Variation of the specific cake resistance (coagulant dose in 0, 1, 3, 5, and 7 mg/L).

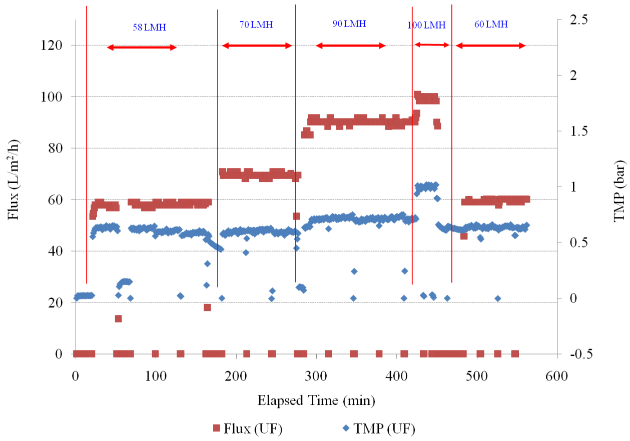

Fig. 6. Determination of critical flux by flux step method (without coagulation).

Table 4
Optimum coagulant condition of jar test

$FeCl_3{\cdot}6H_2O$ concentration (mg/L)	Turbidity (NTU)
0.0	6.03
3.8	0.73
7.6	0.50
11.4	0.76
15.2	0.47
19.0	0.52
22.8	1.04

cake resistance does not always decrease with an increase in coagulant dose. Under the condition at a high coagulant dose and a neutral pH, the density of the flocs is low and

Table 5
Variation of the specific cake resistance (coagulant dose in 0, 1, 3, 5, and 7 mg/L)

$FeCl_3{\cdot}6H_2O$ concentration (mg/L)	Specific cake resistance (m/kg)
0.0	3.23×10^{14}
1.0	2.76×10^{14}
3.0	3.69×10^{14}
5.0	2.03×10^{14}
7.0	2.30×10^{14}

their compressibility is high. Accordingly, the cake is compressed under the application of pressure, leading to high specific cake resistance at high coagulant dose. This is probably the reason why the specific cake resistance is the lowest at 5 mg/L, which is the optimum dose of the coagulant.

3.4. Flux and TMP trends of UF with and without coagulation

The critical flux of the UF for the feed water without coagulation was measured by the flux step method as shown in Fig. 6. With the stepwise increase in the flux, the TMP increased. The critical flux was determined at the flux condition where the TMP became unstable and suddenly increased. Based on the results in Fig. 6, the critical flux seems to be approximately 100 L/m² h.

The flux step method was repeated to measure the critical flux of the UF for the feed water with the coagulant dose of 5 mg/L. Fig. 7 shows the experimental results for the determination of the critical flux. As the flux increases from 60 to 160 L/m² h, the TMP increased. However, there was no flux condition to rapidly increase TMP. This suggests that the critical flux for the feed water with the coagulation is higher than

160 L/m² h. A more stable operation of UF can be expected for this feed water at high flux condition compared with the feed water without coagulation.

After the critical flux measurement, a set of continuous UF operations were carried out. Fig. 8 shows the variations of flux and TMP during the continuous operation of UF using the feed water without coagulation. The flux was 60 L/m² h and the initial TMP was 0.7 bar. After 30 d, the TMP began to increase and reached to 1.5 bar. Then, the CIP was performed and the UF operation was resumed. Even though the operation flux was lower than the critical flux, the TMP was found to increase. This suggests that the critical flux cannot explain UF membrane fouling during long-term operations. Moreover, the results imply that the feed water without coagulation has substantial fouling potential to make the TMP unstable in the UF process.

Fig. 9 shows the changes in flux and TMP during the continuous operation of UF using the feed water with the coagulant dose of 5 mg/L. Initially, the UF was operated at 60 L/m² h and little increase in TMP was observed. Since the critical flux of this feed water was above 160 L/m² h, the flux was increased to 90 L/m² h after 3 months operation. Again, the TMP was maintained stable and the fouling was negligible. It is evident from the results that the coagulation is effective to retard UF fouling. It also allows the UF operation at higher flux without the needs of frequent CIP.

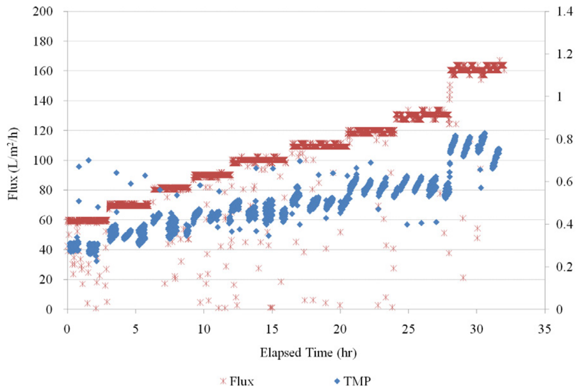

Fig. 7. Determination of critical flux by flux step method (with coagulation).

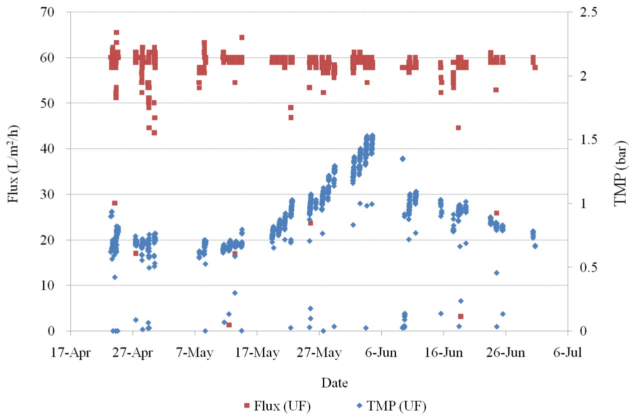

Fig. 8. The variation of TMP in continuous operation mode (without coagulation).

3.5. Chemical cleaning

When the TMP reached to the maximum pressure (1.0–1.5 bar), chemical cleaning was performed. Table 6 summarizes the procedures for CIP. First, the pure water flux of fouled membrane was examined to determine the TMP before CIP. Then, the membrane was contacted with cleaning chemical solutions and rinsed. Table 7 presents the recovery ratios of TMP at different CIP conditions. The alkali/acid cleaning significantly improved TMP recovery to 92% whereas the alkali cleaning did not show high TMP recovery. The acid cleaning resulted in the TMP recovery of 82%, which is higher than that of the alkali cleaning but lower than that of the alkali/acid cleaning. These results suggested that the foulants contains not only inorganic particles but also organic matters. Accordingly, the combination of NaOH and citric acid was more effective to recover TMP than the single cleaning solutions such as NaOH or H_2SO_4. It is likely that the inorganic foulants are removed by the citric acid and the organic foulants are removed by the NaOH solution.

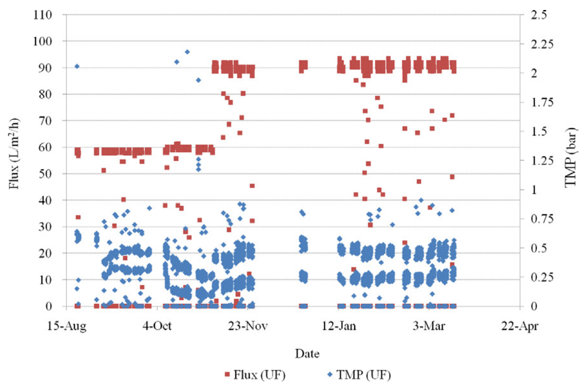

Fig. 9. The variation of TMP in continuous operation mode (with coagulation).

Table 6
Operation condition on CIP

CIP methods	Cleaning chemical	pH	Cleaning time
Initial	–	–	3 h (30°C)
Alkali cleaning	NaOH	12.5	3 h (30°C)
Acid cleaning	H_2SO_4	2.5	3 h (30°C)
Alkali/acid cleaning	NaOH + citric acid	NaOH: 12.5	3 h (30°C)
		Citric acid: 2.5	2 h (25°C)

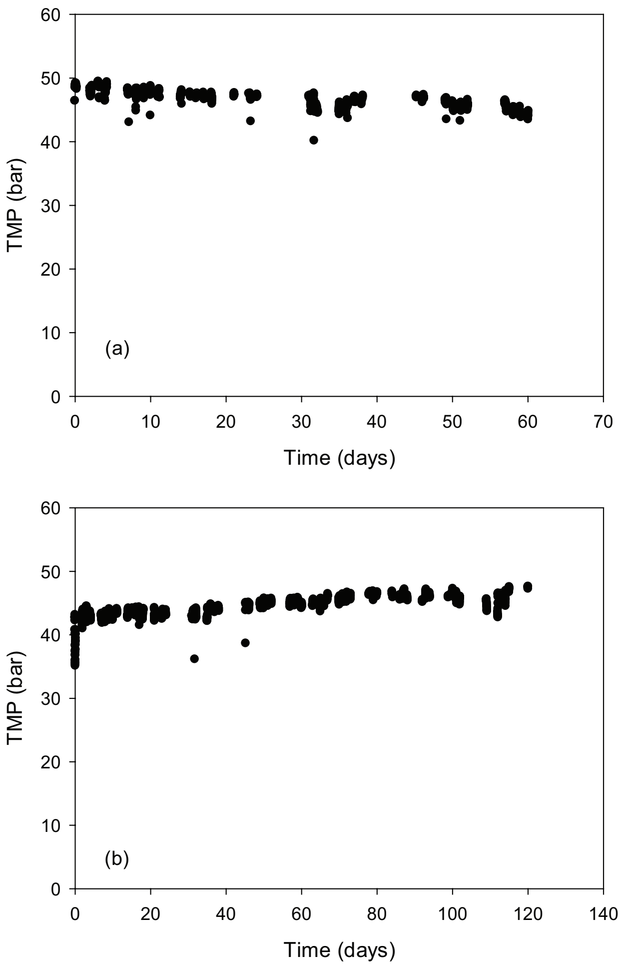

Fig. 10. Changes in TMP during SWRO operation using UF pretreatment and coagulation–UF pretreatment. (a) UF pretreatment and (b) coagulation + UF pretreatment.

3.6. Effect of UF pretreatment on SWRO performance

Both UF and coagulation–UF produced water with high quality as RO feed water. The turbidity of the raw seawater ranged from 8 to 70 NTU and that of the pretreated seawater by the UF or coagulation–UF ranged from 0.1 to 0.5 NTU. The SDI of the pretreated seawater was less than 3.5 in most cases. Accordingly, the TMP of the seawater reverse osmosis (SWRO) was stable with the UF or coagulation–UF pretreatment as illustrated in Fig. 10. Since additional removal of organic matters can be done by coagulation with UF, the TMP of SWRO after the coagulation–UF pretreatment was slightly higher than that after the UF pretreatment. Fig. 11 compares the TMP values for the two cases. The average TMP values of the SWRO for the UF and coagulation–UF were 46.7 and 44.5 bar, respectively. This suggests that the combination of coagulation with UF is beneficial not only for UF fouling control but also for SWRO fouling prevention. Nevertheless, the use of coagulant may increase the operation cost and should be applied by considering the trade-off relationships between the reduced energy consumption and the increased chemical cost.

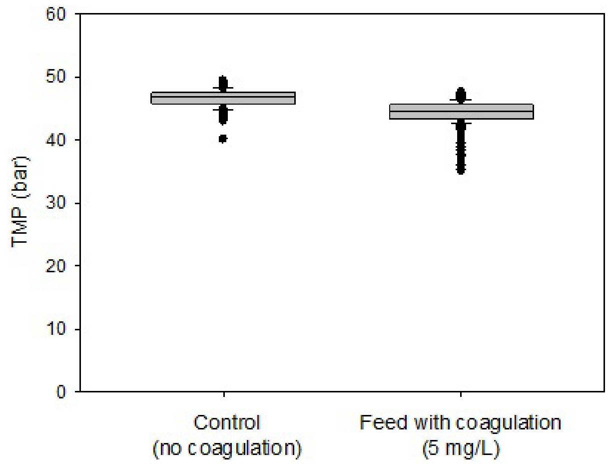

Fig. 11. Comparison of TMP for SWRO operation with UF pretreatment and coagulation–UF pretreatment.

Table 7
Effect of CIP method on TMP recovery ratio

CIP methods	TMP (bar)	Recovery (%)
Initial	0.55	100
Alkali cleaning	0.89	62
Acid cleaning	0.67	82
Alkali/acid cleaning	0.60	92

4. Conclusion

This study provides the opportunity to compare performance of UF membrane filtration with and without coagulation. From the experimental results obtained, the following conclusions can be drawn:

- The optimum coagulant ($FeCl_3$) dose for the seawater used in this study was 5 mg/L. Not only the turbidity of the supernatant in the jar tests but also the specific cake resistance was low at this coagulant dose.
- Although UF itself could be directly applied for the pretreatment of seawater, fouling occurred to decrease the reduced flux (J/J_0) in the short-term tests and increase the TMP in the long-term operation. Use of coagulant together with the UF was found to be effective to retard fouling.
- The critical flux for the feed water without coagulation was determined to be 100 L/m^2 h while that for the feed water with coagulation was higher than 160 L/m^2 h. This allows the continuous UF operation not only at a low flux condition (60 L/m^2 h) but also at a high flux condition (90 L/m^2 h).
- The alkali/acid cleaning significantly improved TMP recovery to 92% whereas the alkali cleaning and acid cleaning showed insufficient TMP recovery. This is attributed to the existence of organic and inorganic foulants on the UF membrane.
- The TMP of SWRO after the coagulation–UF pretreatment was slightly higher than that after the UF pretreatment.

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