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Evaluation of different cleaning strategies for different types of forward osmosis membrane fouling and scaling

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Abstract

In this study, various cleaning methods are investigated to effectively control membrane fouling (i.e., organic fouling, colloidal fouling and scaling) in forward osmosis (FO). Simple physical cleanings (i.e., hydraulic flushing and osmotic backwashing) are firstly employed to remove membrane fouling. Both methods achieved higher than 95% efficiency and osmotic backwashing has a better efficiency for organic fouling and scaling in long-term operation, but colloidal fouling was not removed. To enhance the efficiency, intense physical cleanings (i.e., air scouring and the combination of various physical cleaning) and chemical cleanings (i.e., EDTA, low pH and high pH) were conducted but could not remove the fouling layer possibly because of the polymerization of silica colloids. Lastly, four hybrid mitigation methods (i.e., low pH DS, pulsed flow, high crossflow, and periodic sparging of CO₂ saturated solution) followed by hydraulic flushing were examined. The combination of CO₂ saturated solution and hydraulic flushing completely removed the fouling layer because CO₂ bubbles weakened the interaction between the fouling layer and the membrane surface. Periodic air scouring also exhibited similar effect on the fouling layer. Therefore, periodic air scouring can be more feasible option to control the silica colloidal fouling but not organic fouling.

Keywords: Forward osmosis; Colloidal organic and silica fouling; Membrane cleaning; Fouling control; CO₂ nucleation; Air scouring.

1. Introduction

Forward osmosis (FO) has been recently getting attractions as an alternative or a supplement to other membrane-based desalting processes such as nanofiltration (NF), reverse osmosis (RO) and membrane distillation (MD) [1-3]. In FO, a highly concentrated draw solution (DS) is utilized to induce the osmotic pressure difference as a driving force and thus extract water molecules across the semi-permeable membrane. Due to low hydraulic pressure and utilization of the DS, FO can have many beneficial properties such as low-energy requirement, high fouling reversibility and high recovery rate [4]. Therefore, FO has been intensively studied for treating wastewater having high fouling potential such as municipal wastewater [5-7], shale gas and coal seam gas (CSG) produced water [8, 9], industrial wastewater [10, 11], and landfill leachate [12].

Many studies have been carried out to investigate the fouling reversibility in FO. Lee et al. compared the fouling behaviors in FO and RO and examined their fouling reversibility. High crossflow velocity perfectly recovered the water flux during organic fouling in FO, while no water flux recovery was observed in RO [13]. Mi and Elimelech evaluated organic fouling in FO and RO and demonstrated that organic fouling in FO can be readily removed by hydraulic flushing [14]. They also investigated the mechanism of silica fouling in FO and RO [15] and found out that FO has better fouling reversibility particularly with cellulose triacetate (CTA) FO membrane. However, the combined fouling of organic and colloids showed much lower reversibility than organic fouling and colloidal fouling, respectively, particularly with calcium ions in the feed solution (FS) [16]. When treating the effluent of anaerobic membrane bioreactor (AnMBR) by FO with a fertilizer DS, significant scaling was observed but it was readily removed by hydraulic flushing [17].

Despite high fouling reversibility of FO, irreversible membrane fouling has been also reported by many researchers. Thin-film composite (TFC) polyamide (PA) FO membrane exhibited not only severer organic fouling but also lower fouling reversibility than CTA FO membrane due to the different physicochemical surface property [18]. Both FO membranes

showed similar flux decline but flux recovery of CTA FO membrane was much higher [15]. Flue gas desulfurization (FGD) wastewater was treated by TFC PA membrane and severe membrane scaling occurred. The efficiency of hydraulic flushing was very low because of a thick fouling layer formed on the surface of FO membrane [10]. When treating radioactive medical liquid waste by FO, hydraulic flushing exhibited low efficiency because of the combined fouling of organics and colloids [19]. Treatment of CSG RO brine was also examined using TFC PA FO membrane [8]. The efficiency of hydraulic flushing was very restricted due to CaCO_3 scaling formation.

Furthermore, when using real wastewater, CTA FO membrane has also shown irreversible fouling. When treating real secondary effluent, water flux was greatly restored by hydraulic flushing, but biopolymer-like substances remained on FO membrane surface even after consecutive hydraulic flushing, which requires appropriate pretreatment for the stable operation in the long term [20]. During treating the effluent of a membrane bioreactor (MBR) for treatment of landfill leachate (LFL), less than 90% of water flux was recovered after hydraulic flushing due to polysaccharides and proteins in the fouling layer but chemical cleaning achieved 98.9% of water flux [21]. In addition, membrane orientation has a significant impact on fouling reversibility due to a highly porous structure of the support layer [17, 22].

Therefore, a variety of physical cleaning methods have been investigated in many studies to control and mitigate membrane fouling in FO. As mentioned above, hydraulic flushing (i.e., applying higher crossflow velocity) has been mostly employed to clean the fouled membrane surface due to reversible fouling of FO [10, 19, 23, 24]. Osmotic backwashing (i.e., applying FS having higher osmotic pressure than DS) has been also examined to remove irreversible fouling [8, 17, 25-27]. Some literature shows that osmotic backwashing is effective for colloidal fouling and the combined organic and colloidal fouling in CTA FO membrane [16, 28]. The combination of hydraulic flushing and osmotic backwashing was carried out during the operation of a side-stream anaerobic osmotic MBR [6]. High crossflow velocity and pulsed feed flow could be helpful in mitigating membrane fouling [29]. Airscouring was utilized to effectively control membrane fouling during wastewater treatment by FO [25]. Ultrasound and ultrasonic bath were evaluated for dewatering activated sludge [30] and pre-concentration of high suspended solid content solutions [31]. They could enhance the cleaning efficiency but not completely recover

water flux in some cases. Therefore, various chemical cleaning methods have been proposed and investigated. Wang et al. proposed a chemical cleaning protocol, (i) a mixture of 0.1% NaOH and 0.1% sodium dodecyl sulfate (SDS) and (ii) a solution of either 0.5% HCl or 2% citric acid for municipal wastewater treatment using TFC PA FO membrane [32]. Kim et al. suggested 5% citric acid as a chemical agent for efficient treatment of CSG produced water by FO [8]. Lv et al. evaluated the concentration of actual oily wastewater by FO and examined several physical and chemical cleanings [33]. Despite these efforts, however, there has been no systematic study to investigate membrane fouling and the corresponding cleaning strategies particularly for TFC PA FO membranes.

Therefore, this study aims to systematically explore the cleaning strategies for different types of membrane fouling in TFC PA FO membrane because the CTA FO membrane shows higher cleaning efficiency [15, 16, 28, 29]. Three different types of membrane fouling (i.e., organic fouling, colloidal fouling and scaling) are firstly investigated. Then, simple physical washing methods (i.e., hydraulic flushing and osmotic backwashing) are employed to remove the fouling layer for both short-term and long-term experiments because other studies were conducted for short-term to validate the efficiency of the physical washing methods [34]. Afterward, different types of physical and chemical cleaning methods are examined. Lastly, different hybrid fouling mitigating methods are proposed and investigated. Particularly, CO₂ saturated solution is evaluated as a novel cleaning agent for the first time for FO since it could effectively control membrane fouling in reverse electrodialysis (RED) [35], and ultrafiltration (UF) [36].

2. Materials and methods

2.1 FO membrane

TFC PA FO membrane module (TCK FO 4040) was purchased from Toray Chemical Korea (South Korea). Membrane coupons were collected from the FO module and stored in deionized (DI) water at 4 °C. The detailed information of this membrane can be found elsewhere [8].

2.2 Representative foulants

In this study, alginate, colloidal silica, and gypsum were employed as representative foulants for organic fouling, colloidal fouling and scaling, respectively. Alginic acid sodium salt (CAS No. 9005-38-3) was received and stock solution (1 g/L) was then prepared by dissolving appropriate amount of alginic acid sodium salt in deionized (DI) water. Colloidal silica with 30 nm diameter (CAS No. 7631-86-9) was received in the form of 40 wt. % suspension in water. For gypsum scaling, CaCl_2 (CAS No. 10035-04-8), Na_2SO_4 (CAS No. 7757-82-6) and NaCl (CAS No. 7647-14-5) chemicals were received in the powder form. All chemicals were provided by Sigma Aldrich, USA.

2.3 Lab-scale FO experimental system

All FO experiments were conducted using a lab-scale FO system consisting of a FO module with two symmetric channels (i.e., 100 mm length, 20 mm width and 3 mm depth for each channel), two gear pumps (Cole-Parmer, USA), two flow meters (Cole-Parmer, USA), a balance (MS204, Mettler Toledo, USA), a magnetic stir plate (Cole-Parmer, USA) and two storage tanks [17] as presented in **Fig. 1**. Membrane coupons were installed between two flow channels. Both FS and DS were kept re-circulated in a closed-loop FO system. The FS tank was continuously mixed using a magnetic stirrer. The DS tank was placed on a balance connected to a computer to record weight changes every minute. Data from the balance was used to calculate the water flux using **Eq. (1)**. Then, the normalized water flux was calculated by dividing water flux by initial water flux, as shown in **Eq. (2)**.

$$J_w = \frac{\Delta V_{DS}}{A_m t} \quad (1)$$

$$J_n = \frac{J_w}{J_0} \quad (2)$$

where J_w and J_0 refer to the measured water flux and the initial water flux ($\text{L m}^{-2} \text{h}^{-1}$), J_n is the normalized water flux, ΔV_{DS} is the change in DS volume (L) during operation, A_m refers to the membrane area (m^2), and t refers to the operation time (h).

Figure 1. Schematic diagram of a laboratory-scale FO system used in the present study.

2.4 Lab-scale FO fouling experiments

For organic and colloidal fouling tests, concentrations of alginate and silica were set to be 100 mg/L and 1 g/L, respectively as reported by similar studies to accelerate membrane fouling [16, 29, 37]. Total ionic strength and calcium concentration were fixed at 50 mM and 1 mM, respectively [16, 38]. For scaling tests, 35 mM CaCl_2 , 20 mM Na_2SO_4 and 19 mM NaCl were prepared to make saturation index (SI) 1.3. NaCl of 3 M concentration was employed as DS to induce the osmotic pressure gradient. Crossflow velocity was set at 8.5 cm/s. Solution temperature was maintained at room temperature (23 ± 0.5 °C). A baseline test was conducted using FS without foulants to evaluate the flux decline by membrane fouling and the resulting flux curves were also utilized to calculate fouling resistance via the modified classical resistance in series model, as shown in **Eq. (3)** [38]. Although this model can be restricted in FO because of a variety of influencing factors (i.e, external concentration polarization, internal concentration polarization, cake-enhanced osmotic pressure, and hydraulic resistance), this model can be very useful to compare and easily quantify the effect of the fouling type on flux decline. Then, specific fouling resistance was obtained as **Eq. (4)**.

$$\frac{J_w^0 - J_w'}{J_w'} = \frac{R_f}{R_m} \quad (3)$$

$$R_f' = \frac{R_f A_m}{m_a} \quad (4)$$

where J_w^0 and J_w' are water fluxes of pristine FO membrane and fouled membrane, respectively ($\text{L m}^{-2} \text{h}^{-1}$), R_f and R_m are the fouling resistance of fouled membrane and pristine FO membrane,

respectively (m^{-1}), R'_f is the specific fouling resistance of fouled membrane (m kg^{-1}), and m_a is the mass of foulants on the membrane surface (kg^{-1}). Flux decline rate during fouling experiments can be obtained using **Eq. (5)**.

$$FDR = \left(\frac{J_{w,i} - J_{w,t}}{J_{w,i}} \right) \left(\frac{1}{\Delta t} \right) \quad (5)$$

where $J_{w,i}$ and $J_{w,t}$ are initial water flux and reduced water flux after time ($\text{L m}^{-2} \text{ h}^{-1}$), respectively and Δt is the operation time (h).

2.5 Membrane fouling cleaning experiments

2.5.1 Physical cleanings

Two physical cleaning methods, i.e., hydraulic flushing and osmotic backwashing, were examined for all fouling experiments. Hydraulic flushing was conducted by flushing DI water inside both flow channels at a high crossflow velocity (25.5 cm s^{-1}) for 15 minutes. For osmotic backwashing, 1M NaCl DS solution and DI water flowed in a FS channel and a DS channel, respectively at 8.5 cm s^{-1} crossflow velocity for 15 minutes to generate reverse water flow to the fouling layer. Physical cleanings were conducted after collecting the accumulated permeate of 0.4 L for the short-term evaluation and after every 24 hours for the long-term evaluation. After cleaning, both FS and DS were freshly replaced with new solutions. Flux recovery rate was calculated by comparing initial water flux before fouling experiments and after physical cleaning. The salt residual can remain in the flow channel and inside the FO membrane. Therefore, DI water was circulated for 15 minutes after osmotic backwashing.

2.5.2 Intense physical cleanings for silica colloidal fouling

Intense physical cleanings were further examined only for silica colloidal fouling. FO experiments were carried out for 24-hours filtration and then followed by 15-minute intense physical cleanings (i.e., air scouring, the combined air scouring and osmotic backwashing, and the combined air scouring, hydraulic flushing and osmotic backwashing). Air scouring was employed by continuously injecting air bubbles for 15 minutes at the same flow rate to that of FS because it has been widely used for intensive cleaning in membrane technologies, such as MBR [39], UF [40], and FO [41, 42]. Then, air scouring and osmotic backwashing were integrated and applied to remove the fouling layer. Lastly, air scouring, hydraulic flushing and osmotic

backwashing were further combined to enhance the efficiency of physical cleaning on silica colloidal fouling. After the intense physical cleaning, experiments were continued for another 24-hour filtration.

2.5.3 *Chemical cleaning for silica colloidal fouling*

Three different chemical cleaning agents (i.e., ethylenediaminetetraacetic acid (EDTA), nitric acid and sodium hydroxide) were adopted in the present study. 5 mM EDTA was examined as a metal chelating agent for the control of inorganic membrane fouling [9]. Sodium hydroxide and nitric acid were utilized to adjust pH of DI water to pH 12 and pH 2, respectively. For chemical cleaning, a cleaning agent was provided inside the FS channel and DI water was flushed inside the DS channel for 15 minutes after 24 hours FO operation.

2.5.4 *Fouling mitigation for silica colloidal fouling*

A variety of fouling mitigation strategies were proposed and examined in this study to control membrane fouling in FO. Firstly, low pH DS (pH 2) was prepared as a candidate for mitigating silica colloidal fouling since the colloidal stability is influenced by the reverse diffusion of hydrogen ions [28]. High reverse diffusion of hydrogen ions can reduce FS pH and then prevent the aggregation of silica colloids owing to the short-range strong repulsive hydration forces at low pH [28]. Then, turbulent flow type and high crossflow velocity (25.5 cm s⁻¹) were adopted since the flow type significantly influences membrane fouling in the membrane-based processes [29, 43]. To induce turbulent flow, a peristaltic pump (Cole-Parmer, USA) was utilized. Lastly, carbon dioxide (CO₂) saturated solution was utilized as a cleaning agent since it can control membrane fouling in RED [35]. CO₂ saturated solution was prepared by dissolving CO₂ gas in DI water under 2 bar and then injected to the module for a second per every hour. In order to elucidate the mechanism of CO₂ saturated solution, air scouring was further examined by injecting air for a second every hour. The flow rate of injected air (0.3 L min⁻¹) was the same to that of FS. After 24 hours operation, hydraulic flushing was conducted to evaluate how much the fouling mitigation methods can influence the efficiency of hydraulic flushing on flux recovery.

2.6 Membrane surface characterization

For membrane surface characterization, membrane coupons were collected after experiments, soaked in DI water to remove residues of both FS and DS, and then dried in a desiccator for 1 day. The surface morphologies of pristine and fouled FO membranes were analyzed by scanning electron microscopy (SEM, Zeiss Merlin, Carl Zeiss AG, Germany) and energy dispersive X-ray spectroscopy (EDX) at an accelerating voltage of 3 kV and 20 kV, respectively. Membrane surface roughness was analyzed by atomic force microscopy (AFM, Dimension Icon, Bruker, Germany) under ambient conditions in non-contact mode with silicon probes (FESPA-V2, Bruker, Germany). Turbidity was measured by 2100AN Turbidimeter (Hach, USA).

3. Results and discussion

3.1 Membrane fouling and physical cleaning

3.1.1 Membrane fouling behavior

To evaluate membrane fouling (i.e., organic fouling, colloidal fouling and scaling) in FO, FO experiments were conducted using alginate, silica and gypsum as representative foulants. SEM images (**Fig. 2** and **Fig. S1**) indicate that the membrane surface is completely covered (100% fouling coverage) by organic fouling, colloidal fouling and scaling, respectively, which is confirmed by EDX results (**Fig. S2, supplementary document**). Resulting flux decline curves are normalized and presented as a function of recovery rate in **Fig. 3a**. Recovery rate (%) was calculated by dividing the permeate volume by the initial FS volume. Organic fouling exhibited the highest flux decline followed by colloidal fouling and scaling until 15% recovery rate regardless of their different initial water fluxes (i.e., $25 \text{ L m}^{-2} \text{ h}^{-1}$, $26.9 \text{ L/m}^2/\text{h}$ and $21.9 \text{ L m}^{-2} \text{ h}^{-1}$ for organic fouling, colloidal fouling and scaling, respectively). Afterward, water flux with organic fouling and colloidal fouling was gradually decreased while scaling exhibited more rapid flux decline reaching 4% of initial water flux.

These flux declines could be attributed to increased hydraulic resistance induced by the fouling layer. Hence, we calculated the fouling resistance via the modified classical resistance in series model. **Fig. 3b** shows that the hydraulic resistance of scaling ($79.6 \times 10^{14} \text{ m}^{-1}$) was much higher than those of organic fouling and colloidal fouling ($1.6 \times 10^{14} \text{ m}^{-1}$ and $0.3 \times 10^{14} \text{ m}^{-1}$,

respectively). Due to the different foulant concentrations used in this study, the specific fouling resistance was further adopted using **Eq. (4)** for a fair comparison. **Fig. 3b** indicates that scaling ($130.1 \times 10^{15} \text{ m kg}^{-1}$) showed the highest specific fouling resistance followed by organics and colloids ($3.6 \times 10^{15} \text{ m kg}^{-1}$ and $0.8 \times 10^{15} \text{ m kg}^{-1}$, respectively).

The high specific fouling resistance of scaling could be attributed to the formation of the thick and dense gypsum scaling layer (**Fig. 2d**) induced by the surface properties of TFC PA membrane. Negatively charged TFC PA membrane can increase Ca^{2+} concentration on the membrane surface through the complex with Ca^{2+} ions. This initiates and accelerates the formation of gypsum scales on TFC PA membrane surface, thereby resulting in severer flux decline in TFC PA membrane than CTA membrane [44, 45]. Organic fouling exhibited the second highest specific fouling resistance possibly because the fouling layer completely covered the membrane surface, as shown in **Fig. 2b**. Besides, the compact and dense fouling layer might be formed due to the cross-linking of organic foulants by divalent cations (Ca^{2+} ions) [13]. Non-porous structure of the fouling layer also increased the fouling resistance. In addition, the absence of hydraulic pressure in FO can cause thicker fouling layer and thus accelerate cake enhanced osmotic pressure (CEOP) [46]. Colloidal fouling exhibited the lowest flux decline similar to the baseline (**Fig. 3a**) despite the compact and dense structure, as shown in **Fig. 2c**. This might be because CEOP is the dominant mechanism of flux decline in FO, which is significantly influenced by thickness of the fouling layer [28]. Therefore, the size of colloids significantly influence the flux decline. In this study, colloids with 30 nm diameter were employed and hence the specific fouling resistance could be very low due to its thin and porous layer, which could act as protective layer of the membrane [47].

Figure 2. SEM images of (a) pristine membrane and fouled membranes with (b) organics (alginate), (c) colloids (silica), (d) scaling (gypsum)

Figure 3. FO experimental results in the short term: (a) Normalized water flux as a function of recovery rate (%), (b) hydraulic resistance of the fouling layer, and (c) water flux recovery rate after physical cleaning. Hydraulic resistance and specific resistance of the fouling layer were calculated based on **Eq. (3)** and **Eq. (4)**.

To remove the fouling layer, two physical cleaning methods (i.e., hydraulic flushing and osmotic backwashing) were firstly evaluated after collecting the accumulated permeate of 0.4 L for organic and colloidal fouling and 0.28 L for scaling. **Fig. 3c** shows that hydraulic flushing achieved 95% of initial water flux for all fouling types. Particularly, organic fouling showed the highest water flux recovery (98.9%) followed by scaling (97.6%) and colloidal fouling (96.8%). To confirm whether the fouling layer was removed or not, SEM analyses were carried out using membrane samples collected after physical cleaning. SEM images (**Figs. S3a, S3c and S3e**) indicate that organic fouling and scaling were readily removed by hydraulic flushing while the silica fouling layer still existed even after hydraulic flushing. Results of **Fig. 3c** show that osmotic backwashing could completely restore water flux. From SEM images (**Figs. S3b and S3f**), we can say that osmotic backwashing is very effective in removing the organic fouling layer and the scaling layer. However, the colloidal fouling layer was not removed (**Fig. S3d**), consistent with the result of hydraulic flushing. It is interesting to note that water flux was completely recovered with colloidal fouling although the fouling layer remains on the surface. This is because the colloidal fouling layer has low fouling resistance as discussed above.

3.1.2 Membrane fouling in long term operation

Even though the previous section showed that physical cleaning can readily control membrane fouling and flux recovery in the short-term operation, the long-term operation can limit the cleaning efficiency. Therefore, we carried out FO experiments for 3 days and conducted hydraulic flushing every 24 hours. The obtained water flux was normalized and presented in **Fig. 4a**. For organic fouling, water flux was almost completely recovered even after the second cleaning. In order to evaluate the efficiency of hydraulic flushing, initial flux decline rate for the initial 10 hours of each cycle was calculated using **Eq. (5)**. **Table S1** shows that initial flux decline rate did not change regardless of FO experimental cycles, which indicates the membrane surface was almost completely restored after hydraulic flushing. **Fig. S4a** also support that hydraulic flushing is very effective in removing organic fouling. Nevertheless, AFM analysis (**Table 1**) shows that roughness was slightly increased from 27.5 nm to 34 nm maybe due to the residue of organic foulants.

Fig. 4a indicates that a cleaning efficiency of scaling decreased after the second cycle as water flux was recovered up to 83% and 78.5% of initial water flux. Some scaling residues were also observed on the membrane surface even after hydraulic flushing, as shown in **Fig. S4e**. Interestingly, the initial flux decline rate of scaling was reduced from $27.25 \times 10^{-6} \text{ s}^{-1}$ to $11.77 \times 10^{-6} \text{ s}^{-1}$ while the initial flux decline rate of other foulants (i.e., organics and colloids) either increased or maintained, as shown in **Table S1**. There are two potential reasons of decreased flux decline rate: (i) a decrease in initial water flux of each cycle due to fouling, and (ii) a change of surface physicochemical properties. Water flux influences concentration polarization in membrane-based desalting processes [17] and hence low water flux can help in mitigating membrane scaling via reducing the ion concentration at the surface. Besides, the surface properties might be possibly changed due to the residue of adsorbed foulants which is supported by increased roughness (from 27.5 nm to 41.5 nm) of cleaned membrane. Therefore, the mechanism of the gypsum scaling formation might be changed from heterogeneous surface crystallization to bulk crystallization. As a result, FS turbidity was remarkably reduced from 1721 to 7.14 NTU, which indicates less scaling formation.

We further examined colloidal fouling and found out that water flux was hardly recovered by hydraulic flushing, as shown in **Fig. 4a**. **Fig. S4c** also supports low efficiency of

hydraulic flushing since the colloidal fouling layer still remained on the membrane surface. This could be attributed to the carboxyl functional groups of the FO membrane [15, 48]. Particularly, strong Si-O binding energy can resist shear force induced by high crossflow velocity [48]. It is interesting to note that initial flux decline rate was increased on the contrary to the results of other foulants as presented in **Table S1**. This might be due to significantly increased surface roughness to 59.55 nm even after hydraulic flushing [15].

Figure 4. Flux-decline curves obtained during FO operation with (a) hydraulic flushing and (b) osmotic backwashing. Membrane cleaning was conducted every 24 h by applying hydraulic flushing and osmotic backwashing, respectively.

Table 1. Mean surface roughness after physical cleaning (i.e., hydraulic flushing HF and osmotic backwashing OB).

	Pristine membrane	Colloids		Organics		Scaling	
		HF	OB	HF	OB	HF	OB
Mean roughness (nm)	27.5 (± 3.1)	59.55 (± 14.9)	105.5 (± 19.3)	34.0 (± 5.9)	29.8 (± 2.1)	41.5 (± 1.2)	35.1 (± 2.9)

In order to enhance the cleaning efficiency in FO, then, we evaluated consecutive osmotic backwashing. In case of organic fouling and scaling, **Fig. 4b** indicates that osmotic backwashing could fully recover water flux and showed better efficiency than hydraulic flushing. **Figs. S4b and S4f** also show more clean surfaces. This result is consistent with other studies [8]. In addition, roughness of the membrane surface was restored almost similarly to that of pristine membrane after osmotic backwashing (i.e., 27.5 nm, 29.8 nm and 35.1 nm for pristine membrane, organic fouling and scaling, respectively). Higher cleaning efficiency of osmotic backwashing implies that reverse flow of water flux can be more effective in breaking strong interaction between the fouling layer and the membrane surface. Besides, osmotic backwashing can induce the salt cleaning effect that sodium ions replace divalent cations and then eliminate the foulant-foulant adhesion and the foulant-membrane adhesion [49, 50].

For colloidal fouling, osmotic backwashing could recover water flux as similar as hydraulic flushing for the first cycle but then its efficiency was reduced (i.e., 59.4% and 47.7% for hydraulic flushing and osmotic backwashing, respectively) (see **Fig. 4b**). Low cleaning efficiency of both physical cleaning methods could be attributed to the polymerized silica layer. Silica colloids interact with carboxyl functional groups of TFC PA membrane and then the polymerization of the silica layer occurs via dehydration. The silica layer is continuously dehydrated and a strong gel layer is formed [15, 43]. In addition, the polymerized silica layer is strongly combined with the membrane surface via Si-O bonds [48]. Consequently, it can be concluded that osmotic backwashing is more effective in controlling organic fouling and scaling

than hydraulic flushing. Nevertheless it also needs to further develop better cleaning methods to effectively control colloidal fouling in FO.

3.2 Evaluation of cleaning strategies for silica colloidal fouling

3.2.1 Physical cleaning strategies

Results of **Figs. 4a and 5a** show that air scouring (79.8%) has better efficiency than hydraulic flushing (63.5%). This is because air scouring can provide more aggressive shear force to the fouled surface [25]. However, its efficiency was too low to recover water flux. In order to enhance the cleaning efficiency, air scouring and osmotic backwashing were conducted at the same time since we expected that osmotic backwashing can make the fouling layer more loose and detached from the membrane surface and then air scouring can flush the fouling layer. **Fig. 5a** also shows that water flux was recovered up to 81.5% of initial water flux, which is equivalent to that of air scouring alone (79.8%). We further combined three physical cleaning methods (i.e., air scouring, hydraulic flushing and osmotic backwashing) and conducted the cleaning experiments. Restored water flux was 83% of initial water flux, implying that all physical cleaning applied in this study have very limited efficiency on the removal of the silica colloidal fouling layer. Therefore, we further investigated chemical cleaning to remove the silica colloidal fouling layer.

Figure 5. Flux recovery with (a) intense physical cleaning (i.e., air scouring, the combined air scouring and osmotic backwashing, and the combined air scouring, hydraulic flushing and osmotic backwashing), and (b) chemical cleaning (i.e., EDTA 5 mM, pH 2 solution, pH 12

solution). Membrane cleaning was conducted every 24 h by applying hydraulic flushing and osmotic backwashing, respectively. AS, HF and OB indicate air scouring, hydraulic flushing, and osmotic backwashing, respectively.

3.2.2 Chemical cleaning strategies

Three chemical cleaning agents (i.e., EDTA 5 mM, nitric acid for acid cleaning, and sodium hydroxide for base cleaning) were evaluated in this study. EDTA 5 mM was firstly examined since EDTA can be strongly combined with divalent cations and hence weaken the fouling layer [51]. However, **Fig. 5b** shows that the efficiency of EDTA (67.2%) was restricted and similar to that of hydraulic flushing (63.5%). This is because the silica colloids combine with the membrane surface via the Si-O bond as discussed above. Therefore, there is no divalent cation interaction between the fouling layer and the membrane surface, which leads to no cleaning effect of EDTA.

Then, we evaluated low pH cleaning (pH 2) to remove the colloidal fouling layer since strong acid has been utilized to remove inorganic foulants in RO [52, 53]. Water flux was restored up to 88.1% from its initial value (see **Fig. 5b**), which was higher than that of EDTA 5 mM. High pH cleaning was further carried out (pH 12). Results show that recovered water flux was 83.4% from its initial value, which is higher than that of EDTA 5 mM but less than that of low pH solution. There are two potential reasons of limited cleaning efficiency: (i) short cleaning time, and (ii) a non-penetrative polymerized fouling layer. For chemical cleaning, concentration and contact time are the dominant factors. Thus, contact time (i.e., 15 minutes) of low and high pH solutions may be not enough to remove the polymerized silica fouling layer. In addition, cleaning agents may not penetrate the polymerized silica layer and hence limit the cleaning efficiency. Therefore, it can be said that the silica colloidal fouling layer is not readily removed by both physical and chemical cleaning methods once the silica fouling layer is polymerized.

3.3 Silica colloidal fouling mitigation strategies

In order to control and mitigate the silica colloidal fouling, four different types of fouling mitigation strategies were proposed and evaluated in this study. The resulting flux curves are normalized and presented in **Fig. 6**. To help in understanding the results, the resulting flux curves

as a function of the permeate volume are also presented in **Fig. S5**. For the comparison, a flux decline curve of FO experiments without silica colloids is presented as a baseline.

3.3.1 Low pH draw solution

It was observed that FS pH was decreased to pH 2 during FO experiments. Results of **Fig. 6** show that low pH DS had the lowest flux decline among the mitigation methods tested in this study. To find out the reason of this low flux decline, SEM images were taken on the fouled membrane. **Fig. S6b** shows that FO membrane is fully covered with the fouling layer. Hence, we further carried out an EDX analysis and found out that Si:O ratio is very low (0.46) compared to the baseline (0.78), as shown in **Table 2**, which indicates that a different form of the silica fouling layer was formed. From the Si:O ratio analysis, it can be estimated that the structure of silica colloids was transformed into $\text{H}_5\text{Si}_2\text{O}_7$ (Si:O ratio of 0.5) due to low pH [48].

Then hydraulic flushing was conducted to remove the fouling layer and further recover water flux. **Fig. 6** shows that high flux recovery rate of 95.6% could be achieved compared to the baseline (63.5%). However, **Fig. S7a** indicates the fouling layer still existed on the membrane surface. This implies that the fouling layer formed under low pH has low fouling resistance.

3.3.2 Pulsed flow

Pulsed flow exhibited the lowest initial flux decline rate for 10 hours but reached similar normalized water flux to that of the baseline (see **Table S3**), which indicates that pulsed flow is only effective for the initial stage of colloidal fouling. **Fig. S6c** also shows severe membrane fouling occurred. Then, we carried out hydraulic flushing in order to evaluate whether water flux can be recovered or not. **Fig. 6** shows that water flux was restored up to 87.1%. When considering similar flux decline with the baseline, the cleaning efficiency was very high. This may be because pulsed flow can create highly turbulent flow, thereby enhancing shear force on the membrane surface and causing more membrane movement [29]. However, **Fig. S7b** shows that very thick and dense fouling layer still remained on the surface of cleaned FO membrane.

3.3.3 High crossflow velocity

High crossflow velocity has been widely applied for cleaning and controlling membrane fouling in membrane-based water treatment processes [13, 17]. Besides, a continuous operation under high crossflow velocity has a potential to prevent severe membrane fouling [29]. In contrast to this expectation, high crossflow velocity exhibited most severe flux decline possibly due to higher water flux ($33.4 \text{ L m}^{-2} \text{ h}^{-1}$) compared to the baseline ($27.24 \text{ L m}^{-2} \text{ h}^{-1}$). High crossflow velocity induces low concentration polarization and thus high effective concentration gradient, resulting in an increase in water flux. High water flux leads to high permeation drag force and hence increase the foulant concentration near the membrane surface, thereby causing a more thick and dense fouling layer, as shown in **Fig. S6d** [54]. Besides, high water flux can accelerate the concentration of FS and the dilution of DS, which leads to significant initial flux decline.

Afterward, hydraulic flushing was conducted to remove the fouling layer. Despite the highest flux decline, high water flux recovery (98.3%) could be achieved. This might be because high crossflow velocity induced high shear force and hence the fouling layer might become loose [54,55]. The results also indicate that high crossflow velocity is more effective in restoring water flux than pulsed flow. Nevertheless, it was observed that foulants were not completely removed. It should be noted that physical methods (i.e., pulsed flow and high crossflow velocity) did not influence the form of silica colloids on the membrane surface, as shown in **Table 2**, which implies that they cannot prevent or mitigate the polymerization of silica colloids.

3.3.4 Periodic sparging of CO_2 saturated solution

Recently, CO_2 saturated solution was proposed as a novel cleaning agent [35]. CO_2 saturated solution can not only release CO_2 bubbles but also cause a pH drop, which helps in controlling membrane fouling. To control silica colloidal fouling, therefore, we evaluated periodic sparging of CO_2 saturated solution. **Fig. 6** shows that water flux was rapidly reduced at the initial stage and then gradually decreased. This rapid flux decline could be explained by the accelerated cake enhanced osmotic pressure (A-CEOP) that reversely diffused draw salts which are captured in the fouling layer and hence reduce the effective osmotic pressure gradient across the active layer of the FO membrane. Nevertheless, the lowest flux decline was observed after 1

day operation. An injection of CO₂ saturated solution can decelerate the concentration of the FS, which results in less reduction of the driving force by FS concentration.

Hydraulic flushing was then applied to clean the membrane surface. Interestingly, water flux was completely restored. Besides, it was observed that the fouling layer on the membrane surface was completely removed, as shown in **Fig. S7d** and **Fig. S8**. FTIR analysis results of cleaned membranes (**Fig. S9**) also support the high cleaning efficiency of the combination of CO₂ saturated solution and hydraulic flushing because of its similar peaks to those of pristine membrane. There are two potential reasons of high cleaning efficiency: (i) the formation of micro-bubbles and (ii) a pH decrease. When the pressure of CO₂ saturated solution releases from 2 bar to 1 bar in the module, nucleated micro-bubbles are formed at the inlet of the module and flow through the flow channel. These bubbles can give high shear force to the fouling layer and disturb the strong interaction between the fouling layer and the membrane surface. In addition, FS pH is decreased up to pH 3 due to CO₂ saturated solution. When CO₂ gas is dissolved in the solution, the liquid form of CO₂ reacts with H₂O molecules as presented in **Eq. (6)**. As a result, the weak carbonic acid (H₂CO₃) is formed and the proton (H⁺) becomes liberated into the water as shown in **Eq. (7)**, which makes solution pH decreasing. This pH drop may be able to stabilize silica colloids and hence reduce the formation of the fouling layer [28].

Unlike low pH DS, however, pH decrease by CO₂ saturated solution did not influence the chemical composition of the silica fouling layer since the Si:O ratio was similar between the baseline and CO₂ saturated solution, as shown in **Table 2**. It should be noted that Moreno et al. reported that CO₂ bubble nucleation occurs on the fouling layer and then CO₂ bubbles captures and removes foulants [35]. In this study, CO₂ bubbles were formed before reaching the module due to the release of the pressure. We raised the pressure in the module but CO₂ bubbles were not generated, which resulted in no mitigation effect.

Figure 6. Normalized water flux as a function of operation time with various mitigation methods for silica colloidal fouling. FO was operated by employing different fouling mitigation methods. Then, hydraulic flushing was carried out for 15 minutes.

Table 2. The Si:O ratios of the fouling layer with different fouling mitigation approaches.

	Baseline	Low pH DS	Pulsed flow	3X CFV	Periodic CO ₂ sparging
Si:O ratio	0.78 (± 0.16)	0.46 (± 0.13)	0.75 (± 0.12)	0.84 (± 0.22)	0.74 (± 0.03)

In order to elucidate the mechanism of periodic CO₂ saturated solution sparging on the fouling layer, FO experiments were conducted under different conditions: (i) CO₂ saturated solution (low pH) as FS (i.e., no bubbles) and (ii) periodic air scouring. When using CO₂ saturated solution as FS, the efficiency of hydraulic flushing was higher than the baseline, as shown in **Fig. 7a**. This indicates that a decrease in FS pH (86.6%) could enhance the efficiency of hydraulic flushing. However, the efficiency was much less than that of periodic CO₂ saturated solution sparging (100%). Thus, we further conducted periodic air scouring during FO

experiments with silica colloidal fouling and found out that 100% water flux recovery could be achieved after hydraulic flushing.

To understand the mechanism of CO₂ bubbles on the fouling layer, water fluxes of CO₂ saturated solution and air scouring were normalized and presented in **Fig. 7b**. They exhibited similar flux decline up to 15 hours operation, which implies that CO₂ bubbles and air bubbles can increase A-CEOP via changing the physical structure of the fouling layer since thickness is the most important governing factor to determine A-CEOP in colloidal fouling [13, 28]. Afterward, air scouring showed severer flux decline than CO₂ saturated solution. The higher flux decline observed in this case could be attributed to the higher FS concentration with air scouring because injected CO₂ saturated solution contributed to dilute the FS concentration. From this comparison, the potential mechanism of the bubbles on the formation of the fouling layer can be proposed as several steps: (i) typical fouling is formed on the surface, (ii) bubbles aggressively lead to the movement of the fouling layer, (iii) first two steps repeat during FO experiments, (iv) the fouling layer finally becomes weakly interacted with the membrane surface. Consequently, it can be concluded that the effect of bubbles is dominant to enhance the efficiency of hydraulic flushing.

Even though both CO₂ saturated solution and air scouring can achieve high flux recovery rate, air scouring can be an optimum mitigation method to control colloidal fouling because it empirically takes at least 1 ~ 2 days to saturate solution with CO₂ gas under 2 bar and CO₂ saturated solution also requires an additional storage tank. Therefore, we carried out FO experiments for 6 days to evaluate the sustainability of periodic air scouring. Average water flux of each cycle was calculated and presented in **Fig. 7c**. Average water flux of the first cycle was 18.6 L m⁻² h⁻¹ and it was rarely changed for 6 days, implying that the combination of periodic air scouring and hydraulic flushing can effectively control severe colloidal fouling which is not readily removed by both physical and chemical cleaning. It should be also noted that CO₂ saturated solution may be still a better option for the combined colloidal and scaling fouling due to the synergistic effect of low pH and bubbles nucleation.

Figure 7. (a) Flux recovery rate after hydraulic flushing under different FO operation condition (i.e., a baseline (normal run), periodic sparging of CO₂ saturated solution, employment of CO₂ saturated solution as FS, periodic air scouring), (b) normalized water flux of periodic sparging of CO₂ saturated solution and periodic air scouring, and (c) average water flux of each cycle under periodic air scouring.

4. Conclusions

In the present study, a variety of cleaning methods are investigated for cleaning membrane fouling in FO. Simple physical cleaning is firstly examined for the cleaning of different types of membrane fouling. Then, aggressive physical cleaning and chemical cleaning are conducted to enhance the cleaning efficiency of the colloidal fouling. Lastly, various fouling

mitigation methods combined with hydraulic flushing are evaluated. The major findings drawn from this study are summarized as follows:

- Osmotic backwashing exhibited higher cleaning efficiency than hydraulic flushing for all types of membrane fouling but its efficiency was limited for the silica colloidal fouling.
- Aggressive physical and chemical cleaning could not recover water flux due to the polymerization of the silica colloidal fouling layer.
- The combination of periodic sparging of CO₂ saturated solution and hydraulic flushing could completely remove the fouling layer and restore water flux.
- CO₂ bubbles could weaken the interaction between the fouling layer and the membrane surface, thereby achieving 100% water flux recovery by the aid of hydraulic flushing.
- Periodic air scouring can be more economic option than periodic sparging of CO₂ saturated solution for the control of silica colloidal fouling.

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Highlights

- Osmotic backwashing effectively controlled organic fouling and scaling in FO
- Physical and chemical cleaning had limited effect on silica colloidal fouling
- CO₂ saturated water followed by hydraulic flushing completely recovered water flux
- CO₂ bubbles reduced the interaction between the fouling layer and FO membrane
- Periodic air scouring is more economic option to control colloidal fouling

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: