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Characteristics of membrane fouling by consecutive chemical cleaning in pressurized ultrafiltration as pre-treatment of seawater desalination

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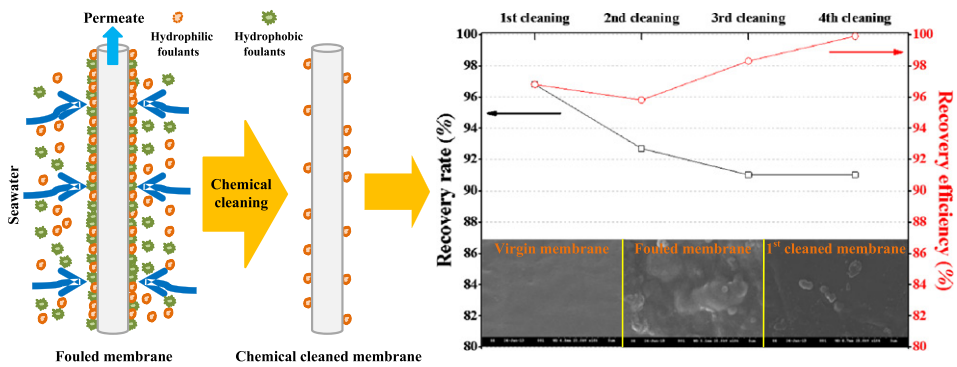
HIGHLIGHTS

- Effect of chemical cleaning on pressurized UF as pre-treatment for desalination
- NaOCl and oxalic acid showed the optimum results for recovering flux rate
- The optimum cleaning combination was oxalic acid–NaOCl–oxalic acid.
- Inorganic salts were found to adhere more on the membrane surface.
- Hydrophobic foulants were removed easily by chemical cleaning on the surface.

GRAPHICAL ABSTRACT



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ABSTRACT

Chemical cleaning of membranes is one of the most important strategies in pressurized hollow fiber ultrafiltration (UF) as a pre-treatment for seawater desalination. Various physical cleaning strategies such as backwashing, aeration and air-scrubbing or chemically enhanced backwashing (CEB) have been investigated in order to remove foulants from the UF membrane. However, the limitation in their cleaning effects being found during long-term operation leads to the need of cleaning in place (CIP) for the recovery of membrane performance. In this study, we used oxalic acid and sodium hypochlorite as chemical cleaning agents. The cleaning in series of oxalic acid–sodium hypochlorite–oxalic acid showed the optimal cleaning efficiency and was applied for the consecutive chemical cleaning. The recovery efficiency of the CIP after first, second, third and fourth cleanings was 96.8%, 95.8%, 98.3% and 99.9%, respectively. It was almost fully recovered to the previous recovered value. However, membrane surface structure was deformed by contact with chemical cleaning agents during cleaning time, because, hydrophilic inorganic foulants are still adhered on the membrane surface even after several cleanings although hydrophobic organic foulants were removed easily by chemical cleaning. An improved CIP strategy should be developed to remove hydrophilic foulants for long-term operation of desalination plants.
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1. Introduction

Nowadays, many regions of the world suffer from the scarcity of fresh water resources for potable, industrial and agricultural purposes. The main problem is the difficulty to supply potable water in water

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shortage areas. Several illnesses are associated with contaminated drinking water. One of the alternative and sustainable ways to produce fresh water is through seawater desalination. Reverse osmosis (RO) is one of the most well-known processes in desalination [1–3]. The RO process is derived from a membrane technology that only allows water to pass through a semi-permeable membrane, and reject the solute (i.e., salt). Seawater is fed to the RO process by using high pressure to gain drinking water [4].

However, seawater cannot be fed directly to RO due to some reasons: first, seawater has inorganic and organic compounds, which can contribute to membrane fouling; and second, if seawater recorded a silt density index (SDI) value of over 5, this could strain the RO membrane. For these reasons, it is necessary to incorporate a pre-treatment method such as coagulation, flocculation, media filtration, multi-media filtration (MMF) and microfiltration/ultrafiltration (MF/UF) in desalination process prior to RO process [5].

Both MF and UF are alternative membrane-based separation methods as pre-treatment process for seawater desalination. There are several advantages in using MF/UF as pre-treatment of RO process. (1) SDI values between 2 and 4 are possible to obtain using this membrane-based pre-treatment, which is more stable compared to other methods; (2) MF/UF is more compact compared to other processes, thus requiring less footprint; (3) MF/UF has a stable flux, and; (4) it can be automated. However, there are also some drawbacks with the use of MF/UF, which include the need for high electrical energy consumption, operating cost and higher initial capital cost [6–9]. Additionally, similar with the RO process, membrane fouling can happen to MF/UF process in a long-term operation, which deters its performance. In the application of MF as pre-treatment for RO, the main foulants are considered to be dissolved solids, suspended solids, organic foulants, big size particles (over 1 μm), etc. On the contrary, small size particles (over 0.04 μm) and organic/inorganic matter such as most of divalent ions and several monovalent ions are thought to be main foulants that can adhere to the UF membrane surface [10,11]. In most reports on chemical cleaning of membranes, researchers usually have focused their attention more on organic fouling rather than inorganic fouling since the membrane processes were designed mainly to remove particle or organic matter from wastewater for meeting regulation of discharge or potable/industrial use. Even though many researches have been investigated on the chemical cleaning of membranes dealing with fresh waters, there is difficulty in application of the results to desalination processes due to the difference in fouling phenomena of membrane filtration treating seawater [12]. To control the fouling, physical cleaning is necessary to be carried out periodically such as backwashing, aeration, air-scrubbing, and chemical enhanced backwashing (CEB). However, physical cleaning and CEB are limited for long-term operation so as not to disrupt the operation [13]. Usually, operation for more than 6 months requires chemical cleaning with various chemical agents. It takes almost one day to perform cleaning in place (CIP) every 6 months of operation.

As you can see in Table 1, many researchers used various chemical agents for cleaning of a wide range of filtration process. The membrane surfaces are exposed to high concentrations of chemical agents for the cleaning process. Different concentrations of chemicals for CIP have been suggested such as 0.5% nitric acid [14], 2% nitric acid [15], 2% sodium hypochlorite, and 1% sodium hydroxide. Kweon et al. [11] used 500 ppm sodium hypochlorite, 250 ppm sodium hydroxide, 2500 ppm citric acid and 250 ppm sodium hypochlorite. Our previous work [16] utilized 0.1%, 0.5%, 1%, 2% and 5% sodium hydroxide in addition to various concentrations (1, 2, and 3%) of nitric acid. However, based from our review of literature, no one has yet investigated the use of chemical cleaning for pressurized hollow fiber ultrafiltration as pre-treatment for desalination by real seawater.

In the present study, fouling of membrane was generated using seawater as feed. The recovery rate was measured after chemical cleaning using various chemical agents at different concentrations, in addition

Table 1
Published reports in literature using different cleaning agents for various processes.

Filtration process	Feed solution	Chemical agents	Membrane material	Reference number
Pressurized MF	Synthetic water (humic acid, Fe, Mn and Ca^{2+})	HNO_3, NaOH	PVDF	[16]
Ceramic MF	3.5 wt.% whey protein	NaOH	Ceramic, zirconium oxide, titanium oxide and aluminum oxide	[14]
Ceramic MF	0.1 g/L of yeast in 10 g/L sugar solution	HNO_3, NaOCl, NaOH	Ceramic	[15]
Submerged MF	Stream water and secondary water from plant	$C_6H_8O_7$, NaOCl, NaOH	PVDF	[11]
Spiral-wound UF	Skimmed milk (11 g/L proteins, 16 g/L lactose and 3 g/L salts)	NaOH, Tween 20, Ultrasil 10	PES	[17]
Submerged UF	Algal-rich water	EDTA, HCl, NaOCl, NaOH	PVC	[18]
Submerged MF and UF	Potable water	$C_6H_8O_7$, NaOCl, NaOH	PVDF and PES	[19]
RO and NF	Licorice aqueous solutions	EDTA, NaOH, HNO_3, H_2SO_4, $CH_3(CH_2)_{10}CH_2OSO_3Na$	Polyamide	[20]
Flat-sheet MF	3.5 g/L of sodium alginate and 2 g/L of BSA	NaOCl	PVDF	[21]
UF	Algae	$C_6H_8O_7$, NaOCl, NaOH	Polysurfone	[22]
UF	Proteins	NaOCl, NH_4OH, machine powder	Modified polysurfone	[23]
UF	Surface water and ground water	$C_6H_8O_7$, NaCl, NaOH, $CH_3(CH_2)_{10}CH_2OSO_3Na$	PES	[24]
UF	Proteins	NaOCl, NaOH, Tween 20	PES and PVDF	[25]
Submerged UF	Surface water	$C_6H_8O_7$, ethanol, NaOH	PVC	[26]
UF	Whey protein isolate	HCl, NaOH	PES	[27]
UF	Surface water	$C_6H_8O_7$, H_2O_2, HCl, Kleen MTC 411, P3 Ultrasil 115, P3 Ultrasil 70, P3 Aquadean Sal, 4AquacleanFer 12	–	[28]
Capillary UF and MF	Reservoir water	$C_6H_8O_7$, H_2O_2, HCl, NaOCl, NaOH	Polyether surfone	[29]
Submerged MF	Micro-polluted raw water	HCl, NaOCl	Polyethlyene	[30]
NF	NOM with ionic compounds	NaOH, $CH_3(CH_2)_{10}CH_2OSO_3Na$	Polyamide	[31]
RO	Alginic acid with 32 g/L of synthetic seawater	De-ionized water, EDTA, NaCl	Polyamide	[32]
Hollow fiber UF	20, 10 and 10 mg/L of humic acid, sodium alginate and BSA	Milli-Q, NaOCl	PVDF	[33]
Submerged hollow fiber UF	Seawater	NaOCl	Polyether surfone	[34]
NF and RO	Oil sand process-affected water	HCl, NaOH	Polyamide	[35]
Flat-sheet MF	1% of milk solution	EDTA	PVDF	[36]
Flat-sheet UF and MF	Whey protein concentrate	NaOH	Polyether surfone	[37]
Flat-sheet MF	Synthetic wastewater	NaOCl	PVDF	[38]

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