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Optimization of chemical cleaning of discarded reverse osmosis membranes for reuse

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Abstract. This study optimized the chemical cleaning process of discarded RO membranes for reuse in less demanding separation processes. The effect of physicochemical parameters, including the temperature, cleaning time, pH of the cleaning solution, and addition of additives, on the cleaning process was investigated. The membrane performance was evaluated by testing the flux recovery rate and salt rejection before and after the cleaning process. High temperatures (45-50 ℃) resulted in a better flux recovery rate of 71% with more than 80% salt rejection. Equal time for acid and base cleaning 3-3 h presented a 72.43% flux recovery rate with salt rejection above 85%. During acid and base cleaning, the best results were achieved at pH values of 3.0 and 12.0, respectively. Moreover, 0.05% concentration of ethylenediaminetetraacetic acid presented 72.3% flux recovery, while 69.2% flux was achieved using sodium dodecyl sulfate with a concentration of 0.5%; both showed >80% salt rejection, indicating no damage to the active layer of the membrane. Conversely, 0.5% concentration of sodium percarbonate showed 83.1% flux recovery and 0.005% concentration of sodium hypochlorite presented 85.2% flux recovery, while a high concentration of these chemicals resulted in oxidation of the membrane that caused a reduction in salt rejection.

Keywords: chemical cleaning; RO membrane; optimization; reuse

1. Introduction

Growing water scarcity around the globe is propelling the water desalination market, which is projected to be valued at over $32 billion by the year 2025 (Adroit Market Research, 2020). Currently, reverse osmosis (RO) technology dominates the global desalination market, accounting for 65% of the installed desalination capacity (García-Pacheco *et al.* 2018). The RO membrane, the "engine" of a desalination plant, is used to produce clean water for both domestic and industrial applications. Performance of the membrane deteriorates because of both irreversible fouling and normal material wear over the lifespan of the membrane, which is typically around 5 to 7 years (Poseidon Water, 2020). The increasing number of RO plants, and the limited lifespan of their membrane modules, are producing an increasing number of discarded membranes, which has become a serious challenge (Lawler, 2015). These membranes are considered to be inert solid waste and are generally disposed of in a landfill because of limited recycling and reuse options (Coutinho de Paula *et al.* 2017). The disposal of used RO membranes is receiving more attention because of the potential economic gains and associated environmental benefits, with recent studies enlightening the importance of reusing discarded RO membranes (García-Pacheco *et al.* 2018) (Coutinho de Paula *et al.* 2017) (de Paula and Amaral, 2018). In addition to extending the membrane lifetime through various optimization methods, the sustainability and efficiency of water treatment plants using RO membrane modules can be further improved if alternatives to disposal, such as reuse and recycling, are considered (Lawler *et al.* 2011). Most commonly, thin-film composite polyamide (TFC-PA)-based membranes are used in RO systems, which comprise a polyamide ultra-thin layer, a supportive microporous polysulfone layer, and a significantly thicker polyester base layer (Lee *et al.* 2011) (Shenvi *et al.* 2015). Chemical cleaning can remove the ultra-thin polyamide layer of the membrane to produce a porous membrane for reuse in other applications (Veza and Rodriguez-Gonzalez, 2003) (Lawler *et al.* 2011) (Pontié, 2015) (García-Pacheco *et al.* 2015). The reuse of the discarded RO membranes in microfiltration or ultrafiltration (UF) applications after chemical cleaning could be a viable alternative to disposal at a large scale.

A literature review summarized the various oxidizing agents, including H_2O_2, $KMnO_4$, NaClO, NaClO+H_2SO_4, and NaClO+SDS (sodium dodecyl sulfate), that have been tested for cleaning, and $KMnO_4$ has proven to be the most effective (Veza and Rodriguez-Gonzalez, 2003). The possibility of reusing the active layer as a UF film was mentioned in previous research (Lawler *et al.* 2013). The spacers and membrane sheets can be used in various geotextile styles through incineration (Mohamedou *et al.* 2010). A pilot-scale validation test of using discarded RO membranes for treating brackish water presented promising results (García-Pacheco *et al.* 2018). An environmental and economic study was conducted for the reuse of the end-of-life RO membrane. Chemical oxidation of thin-film

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composite membranes was performed by immersion in commercial sodium hypochlorite (NaClO) solution. After chemical cleaning, the recycled membrane demonstrated promising results as a replacement for a new ultrafiltration membrane for water treatment with monetary savings of 98.9%. It proved that it is possible to reuse discarded RO membranes and achieve economic gains and environmental benefits (de Paula and Amaral, 2018) (Syed *et al.* 2006).

In view of the literature review, this study aimed to assess the reuse of discarded RO membranes, after chemical cleaning, for application in separation processes with less demanding specifications. Therefore, the optimization of chemical cleaning is of great importance for reuse. Although there have been some previous studies regarding the chemical cleaning of used RO membranes, there are several gaps that should be addressed before large-scale application. The efficiency of chemical cleaning depends on various parameters, including chemical cleaning parameters and the addition of oxidizing agents. Permeability and salt rejection tests measured the effects of the physicochemical parameters. In this study, sodium dodecyl sulfate (SDS), ethylenediaminetetraacetic acid (EDTA), sodium percarbonate, and sodium hypochlorite were added in different concentrations during cleaning to reach an optimum value by simulating a general cleaning-in-place in a laboratory.

Table 1 Characteristics of virgin and discarded RO membranes

Characteristics		Virgin	Discarded
Period of use (y)		-	5
Flux (L/m^2/h) (LMH)		44.47	<20
Salt rejection (%)		94.7	70-90
Membrane features	Type	Thin-film composite	
	Material	Polyamide	
	Element configurations	Spiral-wound	

Table 2 Characteristics of the acid and base cleaning agents

Characteristics	Acid	Base
Phase[(a)]	Liquid	Liquid
pH[(b)]	3.0-3.5	11.5-12.0
Specific gravity[(c)] (g/cm^3)	1.33	1.20
Chemical composition	- Phosphoric acid(H_3PO_4) - Glycolic acid($C_2H_4O_3$) - Citric acid($C_6H_8O_7$)	- Sodium hydroxide (NaOH) - 4Na-EDTA($C_{10}H_{20}N_2Na_4O_{12}$)

(a): At standard temperature and pressure; (b): At 2% concentration in ultrapure water; (c): At 21 °C

2. Materials and methods

2.1 Materials

2.1.1 Membranes

A spiral-wound virgin membrane, and a used RO membrane taken from a filtration plant producing demineralized water from river water situated in Gumi, Korea were used in this study. The discarded RO membrane was manufactured by Toray Co., USA. It had been in operation for 5 years and could not recover its original performance by backwashing and cleaning-in-place (CIP). The general characteristics of the virgin and discarded RO membranes are shown in Table 1.

For the cleaning experiment, the 8-inch (20.32 cm) RO membrane module was disassembled into thin flat membranes and stored in a humid chamber at a constant temperature. For the general experiments, the membranes were immersed in ultrapure water and taken out for each experiment. However, for the discarded RO membrane used in this study, the active layer was already in a dried state; therefore, the membrane was kept dry. The RO membrane was placed in the vessel of a pilot-test device in an elemental state, and a pre-test was carried out with a minimum flow rate of 30% and a salt removal rate of at least 70%. The discarded RO membranes with lower flow rates and salt rejection ratios were not used for this experiment because they were deemed to be deformed or damaged.

2.1.2 Chemical cleaning agents

The commercial cleaning agents used in the experiment were procured from HOIMYUNG Co., Korea. The cleaning agents were used in the general CIP process for RO membranes. They comprised an acid and a base cleaning agent; their characteristics are listed in Table 2.

The detergent improvement experiment was conducted to develop a detergent suitable for cleaning the discarded RO membrane. The additives used to improve the efficiency of the detergent include EDTA, a chelating agent known to be effective in removing membrane fouling in RO membranes, and SDS as a surfactant. Sodium percarbonate ($Na_2CO_3 \cdot 1.5H_2O_2$) and an oxidizing agent capable of damaging the active layer, sodium hypochlorite (NaOCl), were evaluated for possible use in the cleaning process of discarded RO membranes. The manufacturer and chemical characteristics of the selected additives are shown in Table 3.

2.2 Membrane cleaning setup

The discarded RO membrane was collected after 5 years of continuous operation and was used for lab-scale chemical cleaning. A virgin membrane was also selected to compare the performances of the two membranes. The RO membrane modules were transported to our laboratory within 2 h, stored at 4 °C, and examined within 48 h. The characteristics of the RO membrane are presented in Table 1. A lab-scale cross-flow mode setup was used to clean a fragment of the autopsied fouled membrane that was cut (14.6 cm^2) and stored in tap water for cleaning. Stabilization of the membrane was performed by rinsing with ultrapure water for 30 min. The lab-scale experimental setup contained an HP4750 cell (Sterlitech Co., USA) with the specifications that are presented in Table 4.

Table 3 Characteristics of the additives for the cleaning agents

Properties	EDTA	SDS	Sodium hypochlorite	Sodium percarbonate
Formula	$C_{10}H_{14}O_8N_2Na_2 \cdot 2H_2O$	$NaC_{12}H_{25}SO_4$	NaOCl	$Na_2CO_3 \cdot 1.5H_2O_2$
Concentration	98.0%	20% in H_2O	9-11% in H_2O	98.0%
Appearance	Powder	Liquid	Liquid	Powder
Supplier	Daejung Chemicals & Metals Co. (Korea)	Sigma-Aldrich Co. (USA)	Daejung Chemicals &Metals Co. (Korea)	Junsei Chemicals Co. (Japan)
CAS No.	6381-92-6	151-21-3	7681-52-9	15630-89-4

Table 4 Specifications of the HP4750 stirred cell

Active membrane surface (cm^2)	14.6
Maximum pressure (psig)	1000
Maximum temperature (°C)	121
Processing volume (mL)	300
pH range	Membrane dependent
Cell body	316L stainless steel
Cell diameter (cm)	5.1
Cell height (cm)	19.9
Cell width (cm)	14.6

Pre-conditioning is recommended before cleaning or operation by filling the stirred cell with deionized water at a specific temperature and pressure and allowing the cell to operate until stable flux of the permeate is achieved. Afterward, the pressure is released; the water is discarded from the cell and the permeate collection vessel, and then experiments are started by filling the cell with the desired cleaning solution or feed sample; membrane disk should not be allowed to dry. It is a dead-end filtration system that consists of a pressure gas, a feed solution reservoir and stirred hot plate, a permeate tank with a digital balance, a membrane cell, a pressure meter, and a data acquisition computer. The water flux (J_w; LMH) was measured using a digital balance (GF-6100, A&D, USA) and was automatically recorded on a computer. The efficiency of static cleaning was investigated concerning various operating parameters, including temperature, cleaning time, pH of the solution, and the use of combined chemical cleaning agents (such as SDS, EDTA, sodium hypochlorite, and sodium percarbonate).

2.3 Analytical methods

A solution of NaCl with a concentration of 2,000 mg/L was prepared in bulk and stored in a constant temperature and humidity chamber, and the amount required for each experiment was then removed as needed. The filtration pressure was maintained at 15 bar using nitrogen gas, and the filtration was carried out in the total volume. Considering the 20% recovery rate per filtration experiment, the raw water was set to 320 mL, and stirring in the apparatus was set at 3,000 rpm to prevent the concentration polarization from occurring as much as possible. The permeate water produced through the filtration device was collected in a disposable plastic weighing dish on the balance. The amount of this water produced when the filtrate was filtered for 15 min was used to calculate the flux as described in Eq. (1).

$$Flux\ (J_w) = \frac{V_p}{A_m \times T_f} \quad (1)$$

V_p represents the permeation rate (L), A_m is the effective membrane area (m^2), and T_f is the filtration time (h). To compare the cleaning efficiencies according to the cleaning conditions, the flux recovery was computed using Eq. (2).

$$Flux\ recovery\ (\%) = \frac{F_R}{F_V} \times 100 \quad (2)$$

F_R denotes the discarded membrane flux and F_V is the flux of the virgin membrane.

The flux was calculated by measuring the permeate flow rate using the weight of permeate collected in a disposable plastic weighing dish and then transferred to a mass cylinder to measure the electrical conductivity. The electrical conductivity was measured by using an Orion Star A212 (Thermo Fisher Scientific Co., USA). The salt rejection rate calculation was based on the measurement of the electrical conductivity of the raw water and permeate water using Eq. (3).

$$Salt\ rejection\ (\%) = \frac{R_b - R_p}{R_b} \times 100 \quad (3)$$

In this equation R_b and R_p represent the electrical conductivity (μS/cm) of the bulk solution and the permeate water, respectively.

To compare flux recovery and salt rejection rates under various operational conditions with different chemical cleaning agents, an analysis of variance (ANOVA) was used to determine whether there are any statistically significant differences between the results. One-way ANOVA was used in this study, and the statistical analysis of experimental data was performed using R 3.6.3 (The R Foundation, Austria).

3. Results and discussion

The optimization of the chemical cleaning of discarded RO membranes was investigated by considering various operational parameters, such as temperature, cleaning time, and pH of the solution, along with dual and combined

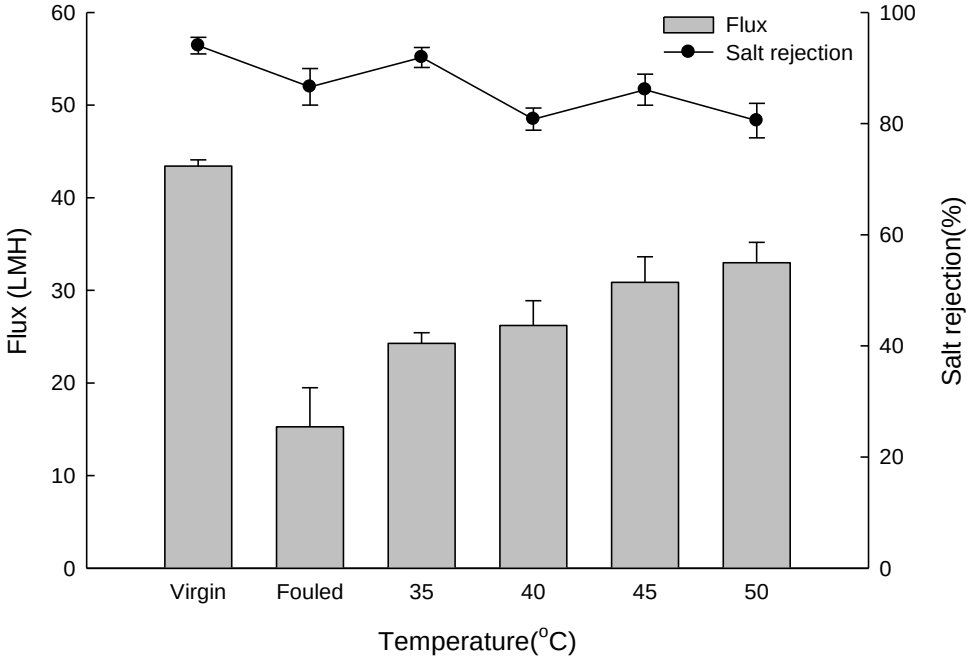


Fig. 1 Effect of temperature on flux and salt rejection

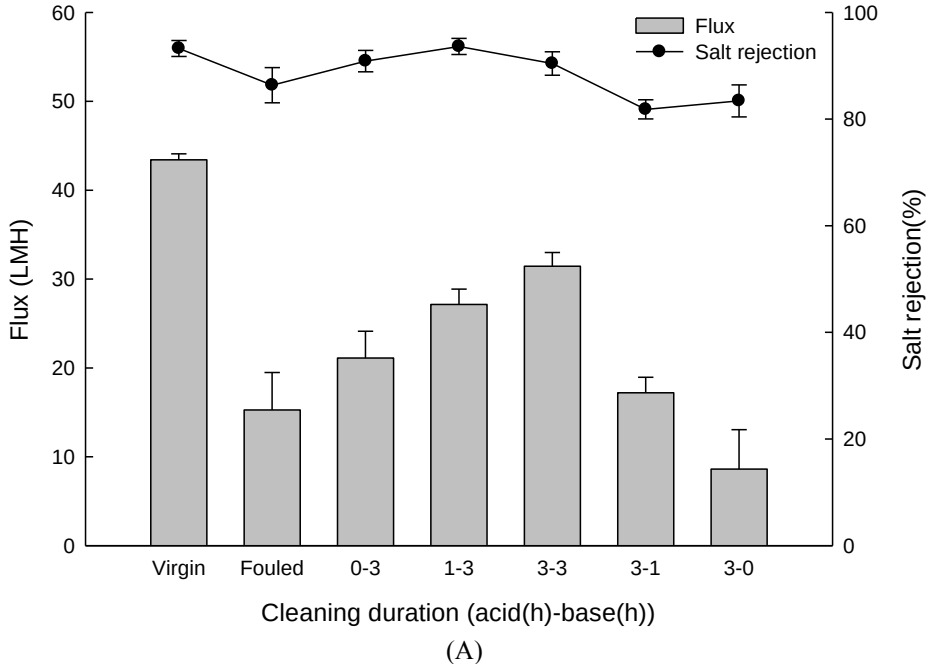
chemical cleaning agents, including SDS, EDTA, sodium hypochlorite, and sodium percarbonate. A standard acid-base cleaning was performed using commercially available acidic and basic cleaning agents, as described in section 2.1.2. Stabilization of the membrane was performed by rinsing with ultrapure water for 30 min before cleaning. Moreover, improvement in cleaning efficiency by the addition of the chemicals was studied. The influence of the operating parameters was explored to find the optimum values for efficient cleaning of discarded RO membranes.

3.1 Effect of temperature

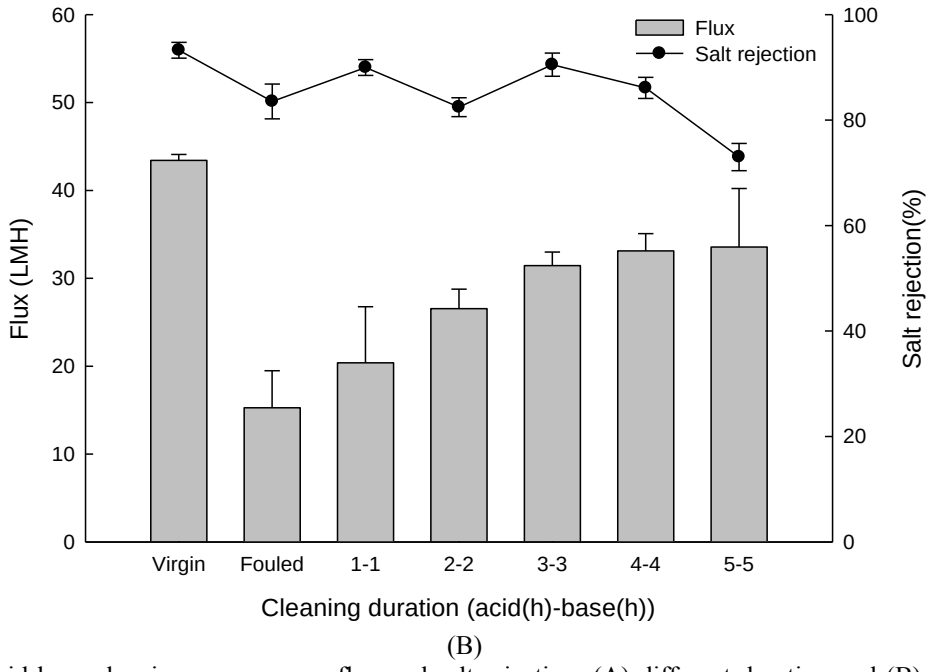
In acid-base cleaning, the effect of temperature on cleaning efficiency was examined, as shown in Fig. 1. The temperature was changed to 35, 40, 45, and 50 °C, while the other parameters were kept constant. The flux recovery rate was measured as 55.9, 60.4, 71.1, and 76.0%, respectively, which indicates that flux increases with temperature. The flux recovery rates over various temperatures were statistically not equivalent ($\alpha = 0.05$). The highest flux recovery rate was observed at 45-50 °C, while the salt rejection rate was more than 80% at all tested temperatures. It was noted that cleaning efficiency increased with increasing temperature from 35 to 50 °C. The diffusive transport of foulants from the membrane surface to the bulk solution and rate of reaction between the cleaning chemical and deposited foulants increased at high temperatures, which resulted in efficient cleaning (Ang, 2008). The high cleaning temperature can enhance the chemical reactivity of a cleaning agent but a very high temperature can damage the membrane, which results in a decrease in salt rejection and a shorter membrane lifespan (Tu *et al.* 2015). In a study of the cleaning of tubular ultrafiltration ceramic membranes, higher temperatures showed a positive impact on cleaning efficiency (Siddiqui and Field, 2016). Therefore, 45 °C was the optimum temperature for the acid-base cleaning process of the discarded RO membrane, with a 71.1% flux recovery rate and >80% salt rejection.

3.2 Effect of cleaning time

The influence of cleaning time on the cleaning efficiency of the acid-base combination was investigated. Fig. 2 summarizes the cleaning efficiency according to the cleaning time for different cleaning orders and combinations. The cleaning times were 0-3, 1-3, 3-3, 3-1, and 3-0, where the first value represents the acid cleaning time (h) and the second one represents the base cleaning time (h), and all other parameters were kept constant. The flux was found to increase with increasing cleaning duration. In Fig. 2(A), the flux recovery rate was 48.63, 62.52, 72.43, 39.61, and 19.82% for cleaning times of 0-3, 1-3, 3-3, 3-1, and 3-0, respectively, with the highest flux recovery and salt rejection at 3-3. Particularly, when the base cleaning time was fixed at 3 h while the acid cleaning duration was changed from 1 to 3 h, the cleaning efficiency improved, and the flux recovery and salt rejection increased in the sequence 0-3 < 1-3 < 3-3. Conversely, when the acid cleaning time was fixed to 3 h and the base cleaning time was varied from 3 to 0 h, both the flux recovery and salt rejection dropped sharply in the order 3-0 < 3-1 < 3-3 compared to the case of the previous order where the acid cleaning time was changed while the base cleaning time was fixed. Therefore, the base cleaning time has a greater effect on the flux recovery and salt rejection than the acid cleaning time.



(A)



(B)

Fig. 2 Effect of acid-base cleaning sequence on flux and salt rejection: (A) different duration and (B) same duration for acid and base cleaning

Another set of cleaning experiment was conducted where the duration of acid and base cleaning was changed in equal amounts between 1-1 and 5-5, as depicted in Fig. 2(B). The flux recovery rate was 46.69, 61.14, 72.43, 76.27, and 77.30% at 1-1, 2-2, 3-3, 4-4, and 5-5, respectively. The highest flux recovery rate was observed at 5-5; however, salt rejection dropped below 80%, and only a 4.87% increase in cleaning efficiency was noticed compared to the case of 3-3. Hence, the most effective cleaning time was 3 h for both the acid and base, which is supported by a previous set of experiments.

The rate of salt removal over time was statistically not equivalent ($\alpha = 0.05$) but not significantly different. It was estimated that the salt removal rates were more than 80% in all cases except acid-base sequence 5-5. Longer contact time will not contribute to an increase in the cleaning efficiency unless there is a favorable chemical reaction between the cleaning agent and foulants in the fouling layer

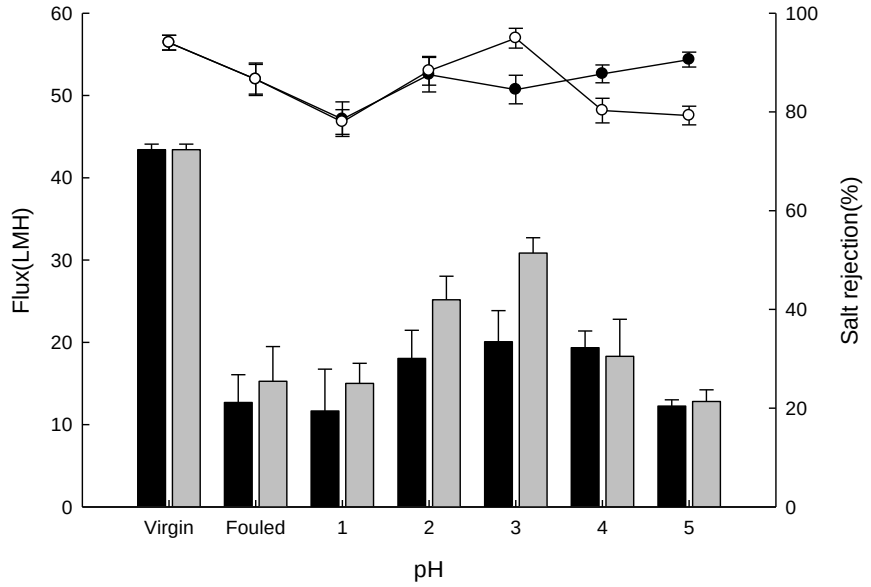


Fig. 3 Effect of pH on flux and salt rejection during acid cleaning

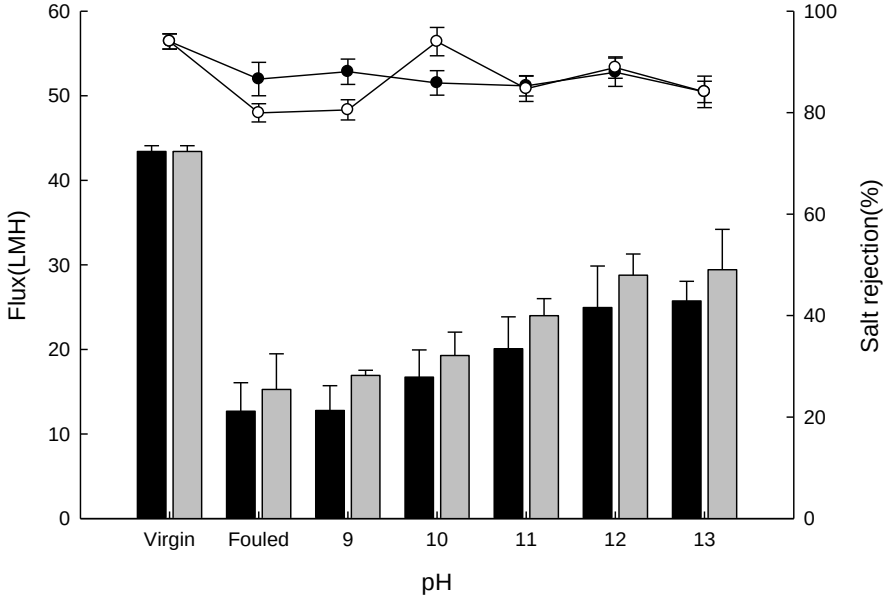


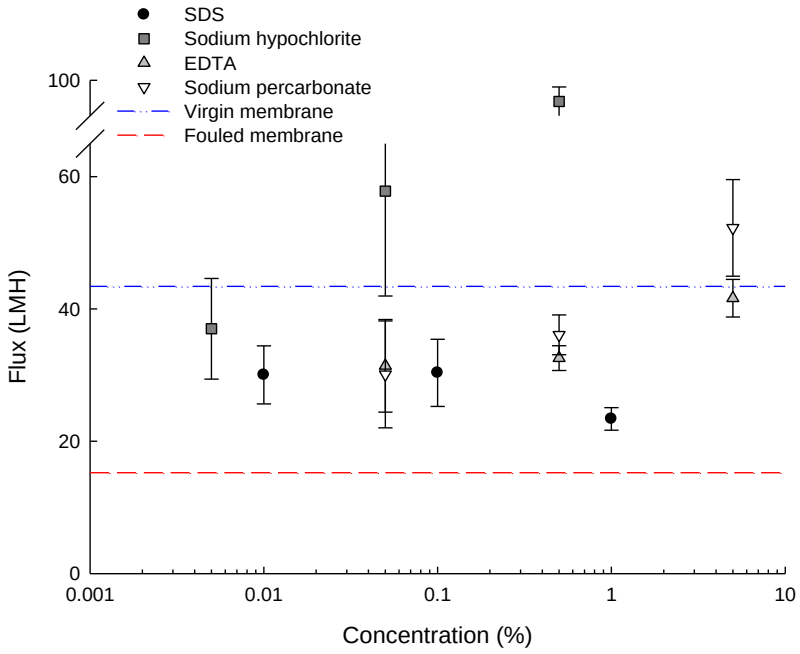
Fig. 4 Effect of pH on flux and salt rejection during base cleaning

to reduce the interactions of the foulant (Ang, 2008) (Ang *et al.* 2011). Therefore, while considering the chemical aspects of the cleaning, optimization of cleaning time should be considered because an increase in cleaning time does not always result in a corresponding increase in cleaning efficiency.

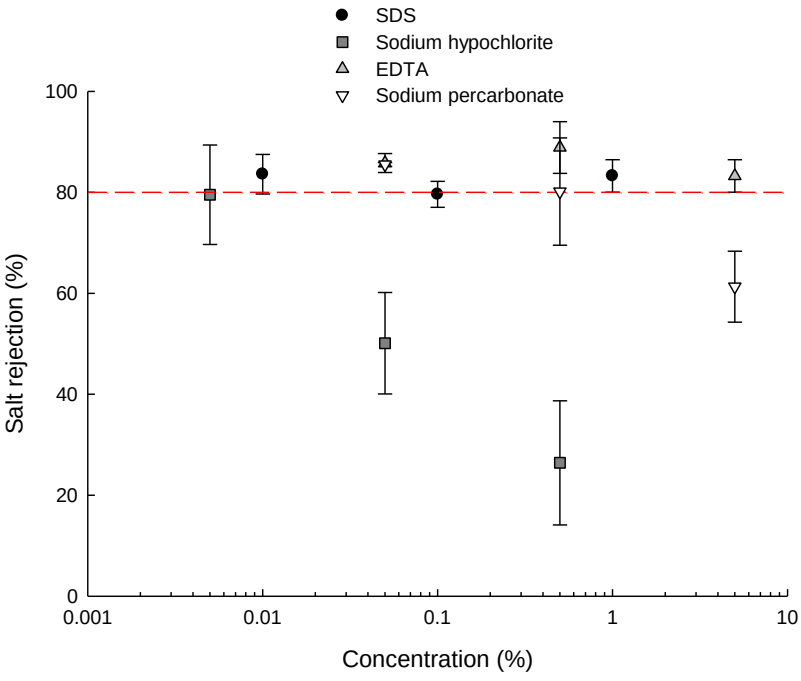
3.3 Effect of pH

The effect of pH on the cleaning efficiency was explored in the acidic and basic range of pH from 1 to 13, as presented in Figs. 3-4, for both organic and inorganic foulants. During acid cleaning, the pH was changed from 1 to 5, while the pH was set to 9, 10, 11, 12, and 13 during base cleaning. The factors other than pH were fixed, and only the change in cleaning efficiency by pH was examined. In addition, the pH of the chemical agent is expected to show different cleaning efficiency depending on the type of foulant in the discarded RO membranes.

Fig. 3 displays that the flux recovery rate of the membrane fouled by organic matter was 34.6, 58.0, 71.1, 24.2, and 29.5 at pH values of 1, 2, 3, 4, and 5, respectively. For inorganic fouled membrane, the flux recovery rate was 26.9, 41.6, 46.2, 44.6, and 28.2% for pH values of 1, 2, 3, 4,



(A)



(B)

Fig. 5 Effect of chemical additives on (A) flux recovery and (B) salt rejection

and 5, respectively. In acid cleaning, both organic and inorganic fouled membranes showed the highest flux recovery rate at pH 3. Within the error range, there was no difference in the flux recovery rate of the membranes fouled by inorganic matter for pH 2-4; the flux recovery rates over pH 2-4 were statistically equivalent ($\alpha = 0.05$). The rate of salt rejection to pH was comparatively better for the removal of inorganic salts (greater than 80%), while for the organic fouled membrane, salt rejection dropped at pH values of 4 and 5 to just 80%. Therefore, there was almost no damage to the active layer of the membrane.

As seen in Fig. 4, the flux recovery rate of the organic fouled membrane was 39.0, 44.4, 55.3, 66.3, and 67.8% at pH values of 9.0, 10.0, 11.0, 12.0, and 13.0, respectively. The flux recovery rate of the inorganic contaminated membrane was 29.4, 38.6, 46.2, 57.5, and 59.3% at pH values of 9.0, 10.0, 11.0, 12.0, and 13.0, respectively. For base cleaning, both organic and inorganic fouled membranes showed the highest flux recovery rate at a pH of 13.0, but when the pH was increased from 12.0 to 13.0, the amount of 1 M NaOH used accounted for 16.7% (50 mL) of the total cleaning solution.

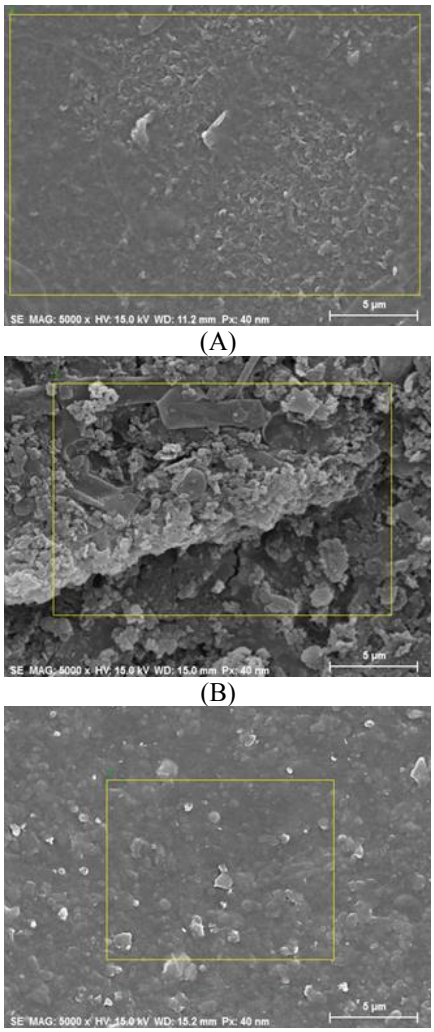


Fig. 6 SEM images of RO membrane: (A) Virgin, (B) Discarded, and (C) Regenerated with optimized cleaning condition

It was determined that the optimum efficiency could be obtained at pH 12 because a slight increase in the flux recovery rate was observed at the maximum consumption cost of NaOH when the pH was maintained at 13. The rate of salt rejection concerning pH was more than 80% regardless of the type of fouled membrane, which indicates that there was almost no damage to the active layer. These results imply that the pH of the cleaning solution is a governing factor affecting the chemical reaction between the acidic or basic cleaning agent and deposited foulants (Ang, 2008), and it can change the surface properties of the membrane; however, the impact of pH on the water permeability can be reversed by applying a subsequent cleaning with an opposite pH condition (Tu *et al.* 2015).

3.4 Effect of chemical additives

Fig. 5 exhibits the effect of chemical additives on the cleaning efficiency of the membranes. The additives, including SDS, EDTA, sodium hypochlorite, and sodium percarbonate, were tested using volume/volume and weight/volume concentrations for liquid and solid additives, respectively. EDTA and sodium percarbonate were used at 0.05, 0.5, and 5%; SDS was used at 0.01, 0.1, and 1%; and sodium hypochlorite was used at 0.005, 0.05, and 0.5%. With the addition of EDTA, the flux recovery rate was 72.3, 75.1, and 95.9% at concentrations of 0.05, 0.5, and 5%, respectively. The highest flux recovery rate was observed with 5% EDTA, but because the consumption was high, 0.05% was considered effective. With the addition of SDS, the flux recovery rate was 69.2, 69.9, and 53.8% at concentrations of 0.01, 0.1, and 1%, respectively. The highest flux recovery rate was obtained when 0.5% was added, but the difference was only 0.71% when compared to the efficiency achieved at 0.01%.

By adding sodium percarbonate, the flux recovery rate was 69.4, 83.1, and 120.37% at concentrations of 0.05, 0.5, and 5%, respectively. The highest flux recovery rate was obtained at the concentration of 5%, but the flux recovery rate was over 100%, and the salt rejection was reduced. Therefore, the concentration of sodium percarbonate was set at 0.5%. The flux recovery rate was 85.2, 133.1%, and 202.8% at concentrations of 0.005, 0.05, and 0.5% sodium hypochlorite, respectively. The highest flux recovery rates were obtained at the concentrations of 0.05% and 0.5%, but the flux recovery rate was over 100%, and the salt rejection was sharply reduced for both concentrations. Consequently, the 0.005% concentration of sodium hypochlorite was considered the optimum value for efficient cleaning.

The concentration of EDTA and SDS did not significantly affect the salt rejections as shown in Fig. 5(B), and it was mostly more than 80%, indicating that there was almost no damage to the active layer of the membrane. In contrast, the addition of sodium percarbonate at a concentration of 5% resulted in the highest flux recovery rate, but the salt rejection declined to 61.32%. Additionally, the highest flux recovery rate was obtained at concentrations of 0.05% and 0.5% of sodium hypochlorite, but the flux recovery rates were over 100% and the salt rejection was dropped to 50.1 and 26.4%, respectively. The oxidation of the active layer of the membrane may cause a flux recovery rate above 100% with lower salt rejection. Therefore, it is important to find an optimum concentration of chemical additives for efficient cleaning of discarded RO membranes to achieve acceptable flux recovery and salt rejection rates for reuse in less demanding separation processes.

The injection of additives for the best cleaning efficiency of the discarded RO membranes used in this study was as follows: (1) Use 0.5% EDTA during the pickling, and (2) mix 0.01% SDS, 0.5% sodium hypochlorite, and 0.005% sodium hypochlorite in the base cleaning. Fig. 6 exhibits the SEM images of the virgin, discarded, and regenerated RO membranes under the optimized cleaning condition. The regenerated membrane was visually the same as a virgin membrane (Fig. 6). Efficient cleaning can be achieved only when both the physical and chemical interactions between the additives and foulants are favorable. During chemical cleaning, a chemical reaction will occur between the cleaning agent and the foulants in the fouling layer. We have demonstrated that the effectiveness of the cleaning process depends on

physicochemical parameters, including the temperature, cleaning time, pH of the solution, and addition of chemical additives in various orders and combinations. The physical parameters, which are primarily responsible for the mass transfer of the reaction products, can play an important role in removing the foulants from the fouling layer. In the cleaning process, favorable chemical reactivity of an additive can enhance the reaction between the additive and the foulants by weakening the structural integrity of the fouling layer (Ang, 2008) (Ang *et al.* 2006).

4. Conclusions

In this study, the cleaning process for discarded RO membranes was optimized to improve the cleaning efficiency for reuse of the membranes in less sensitive separation processes. The optimum cleaning procedure was as follows: rinsing the membrane with ultrapure water for 30 minutes followed by cleaning at a pH of 3.0 for 3 h at 45 °C to remove both organic and inorganic foulants during acid cleaning. In contrast, for the base cleaning process, the best results were achieved at a pH of 12 with the same temperature and cleaning time as that for acid cleaning. It was found that the cleaning efficiency was not significantly different for organic and inorganic foulants. Furthermore, the injection of additives that resulted in the best cleaning efficiency of discarded RO membranes was as follows: (1) Use 0.5% EDTA during the pickling, and (2) mix 0.01% SDS, 0.5% sodium hypochlorite, and 0.005% sodium hypochlorite in the base cleaning. The regenerated membrane had a flux rate of 41.4 LMH and a salt rejection rate of 90.9%. The chemical reaction between cleaning agents and foulants in the fouling layer was greatly influenced by the type and dose of the cleaning agent, as well as the pH of the cleaning solution. The optimized cleaning process can help to reuse discarded RO membranes in less demanding separation applications and can provide economic and environmental benefits.

Acknowledgments

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References

Adroit Market Research (2020), Growing Water Scarcity All Over the World Is Driving the Water Desalination Market Growth; Adroit Market Research, TX, USA. https://www.adroitmarketresearch.com/press-release/global-water-desalination-market-size-and-forecast-2018-2025.

Ang, W.S. (2008), *Optimization of Chemical Cleaning of Organic-Fouled Reverse Osmosis Membranes*, Yale University, USA.

Ang, W.S., Lee, S. and Elimelech, M. (2006), "Chemical and physical aspects of cleaning of organic-fouled reverse osmosis membranes", *J. Membr. Sci.* **272**, 198-210. https://doi.org/10.1016/j.memsci.2005.07.035.

Ang, W.S., Tiraferri, A., Chen, K.L. and Elimelech, M. (2011), "Fouling and cleaning of RO membranes fouled by mixtures of organic foulants simulating wastewater effluent", *J. Membr. Sci.* **376**, 196–206. https://doi.org/10.1016/j.memsci.2011.04.020.

Coutinho de Paula, E., Gomes, J.C.L. and Amaral, M.C.S. (2017), "Recycling of end-of-life reverse osmosis membranes by oxidative treatment: a technical evaluation", *Water Sci. Technol.,* **76**, 605–622. https://doi.org/10.2166/wst.2017.238.

de Paula, E.C. and Amaral, M.C.S. (2018), "Environmental and economic evaluation of end-of-life reverse osmosis membranes recycling by means of chemical conversion", *J. Cleaner Product.,* **194**, 85–93. https://doi.org/10.1016/j.jclepro.2018.05.099.

García-Pacheco, R., Landaburu-Aguirre, J., Molina, S., Rodríguez-Sáez, L., Teli, S.B. and García-Calvo, E. (2015), "Transformation of end-of-life RO membranes into NF and UF membranes: evaluation of membrane performance", *J. Membr. Sci.,* **495**, 305-315. https://doi.org/10.1016/j.memsci.2015.08.025.

García-Pacheco, R., Landaburu-Aguirre, J., Terrero-Rodríguez, P., Campos, E., Molina-Serrano, F., Rabadán, J., Zarzo, D. and García-Calvo, E. (2018), "Validation of recycled membranes for treating brackish water at pilot scale", *Desalination,* **433**, 199–208. https://doi.org/10.1016/j.desal.2017.12.034.

Lawler, W. (2015), "Assessment of end-of-life opportunities for reverse osmosis membranes", Ph.D. Dissertation, School of Chemical Engineering and Faculty of Engineering, USA.

Lawler, W., Antony, A., Cran, M., Duke, M., Leslie, G. and Le-Clech, P. (2013), "Production and characterisation of UF membranes by chemical conversion of used RO membranes", *J. Membr. Sci.* **447**, 203–211. https://doi.org/10.1016/j.memsci.2013.07.015.

Lawler, W., Wijaya, T., Antony, A., Leslie, G., and Le-Clech, P. (2011), "Reuse of reverse osmosis desalination membranes, IDA world congress", *Perth Convention and Exhibition Centre Perth,* Western Australia, USA.

Lee, K.P., Arnot, T.C. and Mattia, D. (2011), "A review of reverse osmosis membrane materials for desalination—Development to date and future potential", *J. Membr. Sci.,* **370**, 1-22. https://doi.org/10.1016/j.memsci.2010.12.036.

Mohamedou, E.O., Suarez, D.B.P., Vince, F., Jaouen, P. and Pontie, M. (2010), "New lives for old reverse osmosis (RO) membranes", *Desalination* **253**, 62-70. https://doi.org/10.1016/j.desal.2009.11.032.

Pontié, M. (2015), "Old RO membranes: Solutions for reuse", *Desalination Water Treat.*, **53**, 1492–1498. https://doi.org/10.1080/19443994.2014.943060.

Poseidon Water (2020), Worldwide Seawater Desalination Capabilities, Poseidon Water, CA, USA. https://www.hbfreshwater.com/desalination-worldwide.html.

Shenvi, S.S., Isloor, A.M. and Ismail, A.F. (2015), "A review on RO membrane technology: Developments and challenges", *Desalination*, **368**, 10–26. https://doi.org/10.1016/j.desal.2014.12.042.

Siddiqui, F.A. and Field, R.W. (2016), "Fouling and cleaning of a tubular ultrafiltration ceramic membrane", *Membr. Water Treat.,* **6**, 141–160. http://dx.doi.org/10.12989/mwt.2016.7.5.433.

Syed, M., Adams, N., Singh, M., Zsirai, I. (2006) "Chemical cleaning for membranes", U.S. Patent Application No. 11/388,648, USA.

Tu, K.L., Chivas, A.R. and Nghiem, L.D. (2015), "Chemical cleaning effects on properties and separation efficiency of an RO membrane" *Membr. Water Treat.,* **6**, 141-160. http://dx.doi.org/10.12989/mwt.2015.6.2.141.

Veza, J.M. and Rodriguez-Gonzalez, J.J. (2003), "Second use for old reverse osmosis membranes: wastewater treatment", *Desalination*, **157**, 65-72. https://doi.org/10.1016/S0011-9164(03)00384-9.