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Evaluating the efficiency of different microfiltration and ultrafiltration membranes used as pretreatment for Red Sea water reverse osmosis desalination

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ABSTRACT

Conventional processes are widely used as pretreatment for reverse osmosis (RO) desalination technology since its development. However, these processes require a large footprint and have some limitation issues such as difficulty to maintain a consistent silt density index, coagulation control at low total suspended solids, and management of higher waste sludge. Recently, there has been a rapid growth in the use of low-pressure membranes as pretreatment for RO systems replacing the conventional processes. However, despite the numerous advantages of using this integrated membrane system mainly providing good and stable water quality to RO membranes, many issues have to be addressed. The primary limitation is membrane fouling which reduces the permeate flux; therefore, higher pumping intensity is required to maintain a consistent volume of product. This paper aims to optimize the permeation flux and cleaning frequency by providing high permeate quality. Different low-pressure polyethersulfone membranes with different pore sizes ranging from 0.1 μm to 50 kDa were tested. Eight different filtration configurations have been applied including the variation of coagulant doses aiming to control membrane fouling. Results showed that all the configurations with/without coagulation, provided permeate with excellent water quality which improves the stability of RO performance. However, more stable fluxes with less-energy consumption were achieved by using the 0.1 μm and 100 kDa membranes with 1 mg/L $FeCl_3$ coagulation. The use of UF membranes, having tight pores, without coagulation also proved to be an excellent option for Red Sea water RO pretreatment.

Keywords: Membrane pretreatment; Microfiltration (MF); Ultrafiltration (UF); Red Sea water reverse osmosis (SWRO); SDI; Membrane cleaning

1. Introduction

Nowadays almost 60% of the feedwater used in desalination plants is seawater and 60% of desalinated water is produced by reverse osmosis (RO) technology (Fig. 1(a)) [1]. This growth in RO market is mainly due to tremendous technology developments making RO technology more economic than thermal-based processes [2]. Over the past 25 years, desalination plants all over the world were moving toward

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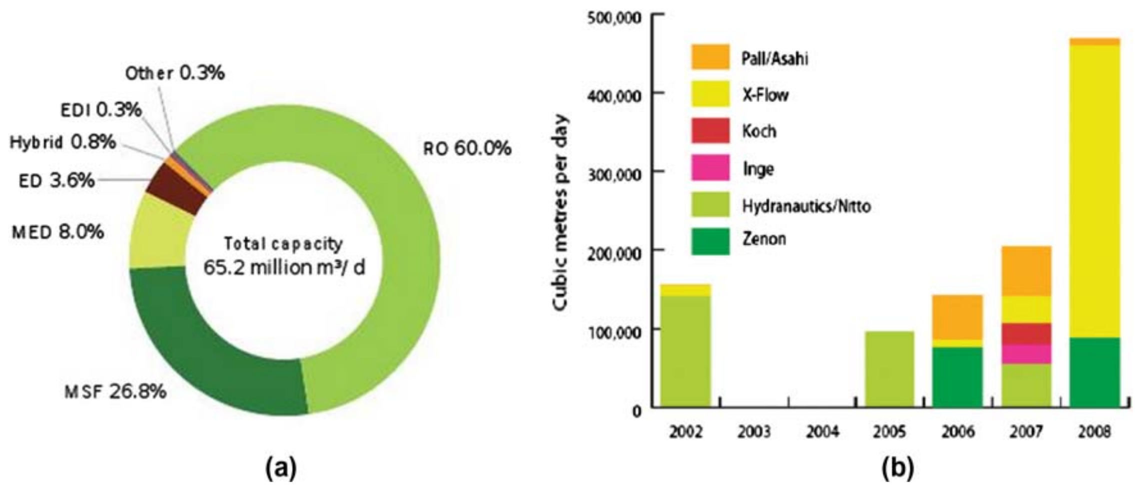

Fig. 1. (a) Global capacity of water desalination by technology [1], (b) UF/MF used in pretreatment for SWRO, annual capacity by manufacturer [4].

membrane technology, because of its advantages compared to thermal processes [3]. The use of low-pressure membranes mainly microfiltration (MF) and ultrafiltration (UF) as pretreatment for seawater RO (SWRO) desalination is increasing as shown by the market trend presented in Fig. 1(b) [4]. Different types of membranes have been developed commercially depending on the process and the type of the treated feedwater quality. The variations include different membrane materials, pore sizes, thickness, module shapes, configurations, and hydrophobicity.

The use of low-pressure membrane technology to produce water with different quality has been widely used for surface water, brackish water, and seawater pretreatment [5]. Proper pretreatment is a key factor for a successful desalination plant operated with RO technology [6–8]. Unlike conventional pretreatment, the use of membrane technology removes particles smaller than the used membrane pore size providing lower silt density index (SDI) values, which makes it an ideal technology for SWRO pretreatment [9]. Only since 2006, the use of MF/UF membranes as pretreatment for SWRO has been globally accepted to replace conventional processes [4]. Several studies showed that CAPEX and OPEX of pretreatment system can reach up to 50% of the total seawater desalination plant cost [10,11]. In brackish water desalination, which represents over 21% of the total desalination capacity [1], the operating cost is lower mainly due to the low energy requirement. However, in remote locations where most of the brackish water desalination plants are installed, sometimes coupled with renewable energy such as solar, wind, and geothermal [12–14], the use of membrane technology may not be suitable due to complexity of its operation [7,8].

A comparison between conventional and low-pressure membranes treatments used in different SWRO pretreatment plants is presented in Tables 1 and 2. From these case studies, the following advantages of using low-pressure membranes can be drawn:

- Higher flux/recovery for RO can be achieved.
- Minimizing the footprint required for the plant.
- Increasing the lifetime of RO membranes, hence reducing the cost.
- Guarantees constant quality of permeate (RO feed) achieved even when raw water quality changes.

This paper focuses on investigating the use of MF and UF membranes as pretreatment of Red Sea water RO. Three polyethersulfone (PES) membranes with different pore sizes (0.1 μm, 100 KDa, and 50 KDa) were used in this study. Experiments were conducted with/without using different $FeCl_3$ coagulant concentrations in dead end filtration mode at both constant flux and constant pressure. The properties and quality of permeate were analyzed in order to identify the best membrane type and configuration.

2. Materials and methods

2.1. Experimental unit

Fig. 2 shows the process flow diagram of the experimental unit that has been designed and used in this study. Red Sea water filtered through a 5 μm cartridge filter to remove large suspended solids is

Table 1
Case studies of SWRO plants using conventional pretreatment process

Ref.	Case name	Raw water source	Source water parameters	Process design	Coagulant	Permeate parameters	Remarks
[15]	Jeddah SWRO plant, 1989	Deep Red seawater disinfected by NaClO	SDI = 5.5–6 and TDS = 43,300 mg/L	Dual media filter (DMF) (anthracite and sand) flowed by 10 μm cartridge filters	0.3 mg/L of $FeCl_3$ mixed with 0.1 mg/L of polyelectrolyte	SDI < 4.0	Coagulants prevent membrane degradation and control quality of the variable SDI
[16]	Doha Research Plant, Kuwait	Surface Arabian Gulf seawater	TDS of 47,000 mg/L and $SDI_{15} > 6.5$	$FeClSO_4$ flocculation and media filter with various grain sizes: silica sand range (0.7–1.2 mm, 1 m high) and anthracite ranges (1.4–2.5 mm, 0.7 m high)	No	SDI = 3.6	Several failures caused by climatic conditions and dosing rate of $FeClSO_4$ and other
[17]	Persian Gulf and Indonesia-1	Persian Gulf	SDI = 6.2, Turbidity 0.22NTU, algae bloom, and hydrocarbon pollution	DAF with double filtration including addition of two coagulants	$FeCl_3$ and polymers	SDI = 1.8–2.9, UV removal = 20–30%	Results in efficient feedwater for RO
[17]	Persian Gulf and Indonesia-2	Persian Gulf	TDS = 25–50 g/L, turbidity 5–20 NTU, pH = 8–8.5	DAF unit followed by polishing horizontal filter	$FeCl_3$ and polymers	Turbidity = 0.25 NTU and SDI < 1.5	The system max. recovery of 35%
[18]	French Institute of Marine Research	Seawater	Turbidity 0.5–4 NTU, TOC = 2.7–6.1 mg/L, and SDI = 6.1–6.4, pH 8, SS = 10–20 mg/L, T = 9–25 °C	Coagulation followed by sand filtration (10 μm)	PAC	SDI = 5.8–5.9	The permeability of the RO membranes has decreased by 28% during 30 days
[6]	ONDEO Services, Gibraltar	Gibraltar seawater	Conductivity = 48.7 mS/cm and SDI = 13–15 and algae bloom	Organic coagulant, three DMF then 10 μm cartridge filters	Organic	SDI = 2.7–3.4	See Table 2
[19]	Singapore SWRO plant	Seawater	SDI = 6.1–6.5, TSS = 6 mg/L	Coarse screens followed by Single Media Filter and 3 stages of 10,5 and 1 μm polishing cartridge filters	Polymeric	SDI = 4 on average	See Table 2
[20]	Ashdod Plant	Mediterranean surface seawater	Turbidity = 1–10 NTU, TDS = 40,500 mg/L, SDI > 6.5, and SS = 2–14 mg/L	Settling and coagulation then media filtration	0.3–0.7 mg/L ferric salt	Turbidity = 0.1–0.2 NTU, SDI = 2.6–3.8	See Table 2

Note: The same colors on the two tables indicate the same research done for both technologies.

Table 2
Case studies of SWRO plants using MF/UF pretreatment process

Ref.	Case name	Raw Water Source	Source Water Parameters	Process Design	Coagulant	Permeate Parameters	Remarks
[18]	French Institute of Marine Research	Seawater	Turbidity 0.5–4 NTU, TOC = 2.7–6.1 mg/L and SDI = 6.1–6.4, pH 8, SS = 10–20 mg/L, T = 9–25 °C	Coagulant and dead end hollow fiber UF membrane (pore size of 0.01 μm)	PAC	SDI = 1–2, turbidity < 0.1 NTU, SS < 0.01 mg/L	Constant permeability for the RO during 20 days trial
[6]	ONDEO Services, Gibraltar	Gibraltar seawater	Conductivity = 48.7 mS/cm and SDI = 13–15 and algae bloom	200 μm pre filter and 100 kDa UF membrane. Then 5 mg/L of free chlorine was added	Organic	SDI < 0.8	Fouling control was more efficient with UF than with conventional pretreatment
[19]	Singapore SWRO plant	Seawater	SDI = 6.1–6.5, TSS = 6 mg/L	Direct flow through 0.1 μm MF membrane flowed by 0.01 μm UF dead end mode	Polymeric	Turbidity = 1.5–3 NTU, TSS = 1.3 mg/L, SDI = 2–3	Flux increased when SDI approx. 3
[20]	Ashdod Plant	Mediterranean surface seawater	Turbidity = 1–10 NTU, TDS = 40,500 mg/L, SDI > 6.5, and SS = 2–14 mg/L	50 μm screen filter and followed by UF filtration using coagulant	0.3 mg/L ferric and 20 mg/L hypochlorite	Turbidity = 0.09–0.16 NTU, SDI = 2.1–3	Seasonal storm increases TSS value then UF becomes preferable
[21]	Addur Desalination Plant	Gulf seawater	SDI = 15–19	Pre-chlorination followed by sand filtration then spiral wound UF membranes	1 mg/L $FeCl_3$ in winter & 2 mg/L in summer		Coagulant improves the filtrate quality

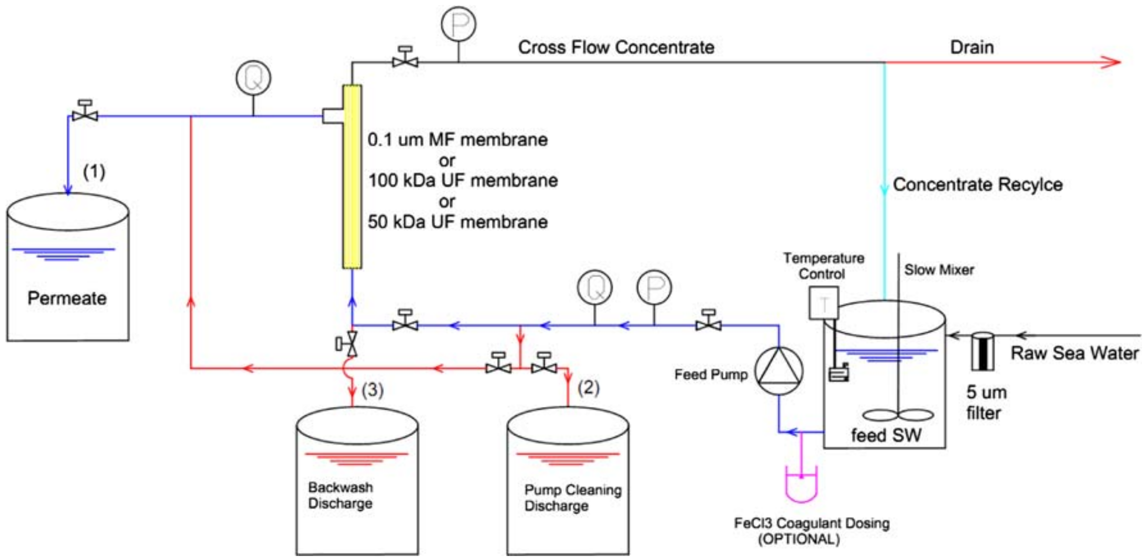

Fig. 2. Experimental unit.

filled in a tank of 75 L and pumped to the MF/UF module. The feed tank was continuously stirred to maintain homogeneous feed quality. Samples of permeate were regularly collected for characterization and analysis. Feedwater temperature was kept constant at 30 ℃ (real seawater temperature during the study period) using a thermoregulator. Different doses of ferric chloride were injected in the feed line. Each experiment was run for 60–65 min. After each experiment, membranes were backwashed using RO permeate for 40–60 s with a flux of about 300 lmh. After each cleaning, membrane permeability was tested to assure the cleaning performance. An additional 40–60 s backwash time in counter flow was performed when the initial membrane permeability was not achieved. Flux measurements and chemical analysis were performed in experiments at constant flux and constant pressure, with and without $FeCl_3$ dosing.

2.2. Water characterization

This research quantifies the performance of MF/UF membranes by testing the quality of permeate mainly SDI, turbidity, pH, conductivity, total dissolved solids (TDS), total suspended solids (TSS), total organic carbon (TOC), natural organic matter (NOM), and temperature (℃).

The SDI testing unit used in this work was Aike Portable Silt Density Index Tester provided by Horizon Environmental Technology Co., Ltd. The laboratory setup measured the rate of plugging of a membrane filter with nominal pore size of 0.45 µm at 30 psi constant pressure filtration for certain period of time (T 5, 10, and 15 min). Turbidity of raw water and permeate product were measured with Hach 2100AN turbidimeter [22]. pH was measured using CyberScan model pH 6,000 m [23]. Both conductivity and TDS of the raw water and permeate product were measured using the equipment manufactured by OAKTON model CON 510 series/Conductivity/TDS/℃/F meter [24]. TSS concentration of Red Sea water was also measured. TSS is quantified using a filtration method described as the TSS dried at 103–105 ℃. The experiment has been conducted according to the standard method ESS Method 340.2: TSS, mass balance (dried at 103–105 ℃) [25]. TOC was measured using TOC-V CPH equipment provided by Shimadzu [26].

2.3. Membranes and process configurations

Hollow fiber modules were used in this study. This type of modules can provide a large-packed membrane area per unit volume, which makes them the most economical and commonly used configuration in SWRO pretreatment [27,28]. The PES fibers (0.1 m^2) in the in-out mode were assembled in a module, where the feed flows inside the fibers and permeate is collected outside [29]. This system has better hydrodynamic flow than the out-in mode [30]. The only difference between the three membranes used in this study is the pore size, which ranges from MF to tight UF (0.1 µm, 100 kDa, and 50 kDa). Table 3 summaries details of the membrane properties used in this investigation.

Table 3
Technical details of the used membranes as given by the manufacturer

Properties	Range
Membrane surface area	0.1 m^2
Flux rate	60–180 lmh
Backwash range	230–300 lmh
TMP for filtration	0.1–1.5 bar
TMP for backwash	0.3–3 bar
Temperature range	0–40 ℃
pH tolerance	1–13
Free chlorine tolerance	Max. 200 mg/L

Table 4
Red Sea water analysis measured during the study period

PH	7.8–8.36
Turbidity (NTU)	0.6–0.75
Conductivity (mS/cm)	60.5
TDS (g/L)	39.8
TSS (mg/L)	0.0,188
SDI_5 (%/min)	11.9
TOC (mg/L)	1.94
DOC (mg/L)	1.17

2.4. Feedwater source

Feedwater was collected from Red Sea, about 2.5 km from King Abdullah University of Science and Technology (KAUST) coastline, located in the western province of Saudi Arabia. The raw water quality analyses are presented in Table 4.

3. Results and discussion

All experiments were conducted during summer. The first experiment was conducted to determine the backwash frequency and operating parameters mainly backwash pressure, flow rate, and time in order to be applied in subsequent experiments. An increase in applied pressure of about 0.2 bars was obtained every 30 min. Fig. 3 shows that membrane cleaning may require more time to reach the initial membrane permeability. Membrane cleaning procedure was described in the previous section.

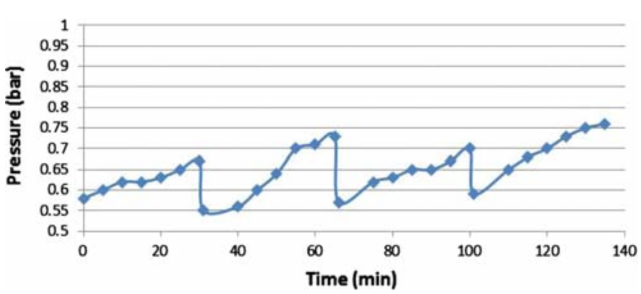

Fig. 3. Backwash frequency test.

3.1. Constant flux experiments

To mimic the industrial operation, constant flux mode experiments were conducted for the three different membranes by injecting different coagulation concentrations. Transmembrane pressure (TMP) of the 0.1 μm MF membrane presented in Fig. 4 shows that the pressure escalation slows down and behaves more steadily with the increase in coagulant concentration meaning that the addition of coagulant enhanced the performance of the membrane. Membrane fouling is likely to be the build-up of a cake on the membrane surface, while without using coagulant smaller particles can penetrate the membrane's pores and block them internally in shorter time [31]. On the other hand, when comparing the different coagulation doses (1, 2, and 3 ppm), it is observed that the 1 mg/L $FeCl_3$ coagulant concentration gives the least increase in pressure with time, which makes it the most preferable option for this membrane. The relative pressure in this configuration shows the minimum growth

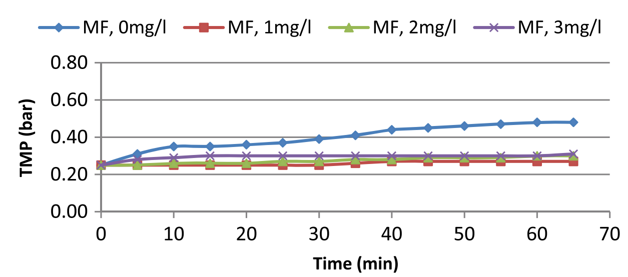

Fig. 4. TMP vs. time, different coagulation doses, 0.1 μm MF membrane at constant flux filtration mode.

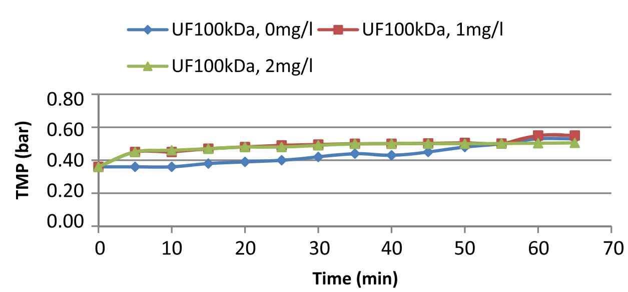

Fig. 5. TMP vs. time, different coagulation doses, 100KDa MF membrane at constant flux filtration mode.

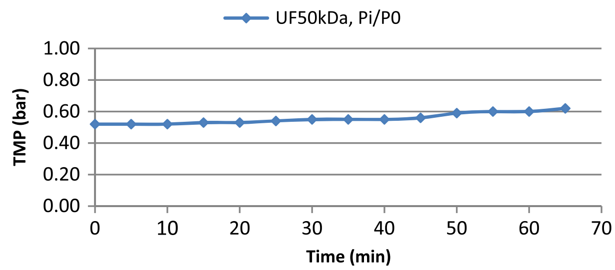

Fig. 6. TMP vs. time, no coagulation, 50 KDa MF membrane at constant flux filtration mode.

making a relative pressure difference of 8% at the end of the filtration period (65 min).

On the other hand, Fig. 5 shows that the performance of the 100 kDa UF membrane at 1 and 2 mg/L $FeCl_3$ injection performed in similar way in terms of $\Delta P_i/P_0$ ratio indicating that 1 mg/L coagulation dose is enough to give the best performance of this membrane. Similarly, TMP vs. time data presented in Fig. 5 shows that TMP increased about 0.1 bar in 50 minutes suggesting the possibility to run without dosing a coagulant. However, coagulation helped in building up a cake on the membrane surface in the first minutes (no significant increase in TMP after 5 min) protecting the membrane from internal blocking.

The 50 kDa UF membrane was used without coagulation as the membrane pores were very tight enabling the removal of small particles present in the feed. In this experiment, higher relative pressure was found ($\Delta P_i/P_0 = 19.2\%$) throughout the same filtration time as the previous experiments (Fig. 6). However, TMP increase was very reasonable despite the need of increasing the membrane cleaning frequency compared to the other two membranes. Therefore, this option is very attractive to be used if the applied environmental regulations are stricter (management of pretreatment sludge disposal), which is the case of several plants such as Perth SWRO desalination plant.

Optimum cases of constant flux experiments for the three membranes are gathered in Table 5. It can be concluded that the 0.1 μm MF membrane with 1 mg/L $FeCl_3$ dosing represents the most suitable and preferred option in terms of cleaning frequency and energy consumption, followed by the 100 kDa UF membrane with 1 mg/L coagulant dose.

Table 5
Optimum $\Delta P_i/P_0$ of the three membranes used at constant flux mode

Membrane	$FeCl_3$ dose	$\Delta P_i/P_0$, %
MF 0.1 μm	1 mg/L $FeCl_3$	8.0
UF 100 kDa	1 mg/L $FeCl_3$	11.3
UF 50 kDa	No coagulant	19.2

3.2. Constant pressure experiments

The aim of this part is to examine the decrease in permeate flux with time while keeping the pressure constant. Data of the 0.1 μm MF membrane presented in Fig. 7 show that the highest relative flux drop ($\Delta J_i/J_0$) was observed for the case without injecting $FeCl_3$. The flux decreased dramatically within 60 minutes (52% decline). However, as the concentration of the coagulant increased from 1 to 3 mg/L, the stability of the flux rate was enhanced and the difference in normalized flux decreased.

Theoretically, the relative flux drop ($\Delta J_i/J_0 = 4.8\%$) with the 3 mg/L was the most preferable option in this case, however, from an economic and environmental point of view, using less coagulant is preferred, especially when the difference in flux decrease between dosing 3 and 1 mg/L coagulant is not high. It is noticeable that the 1 and 2 mg/L $FeCl_3$ had almost the same relative pressure drop. Therefore, by comparing the amount of $FeCl_3$ used and the normalized flux declined in both cases, it was concluded that the 1 mg/L $FeCl_3$ is the preferable option.

In the 100 kDa UF membrane experiments, no coagulant dosing represented the worst case, but it could be the best option in case of chemicals use constraints as the required cleaning frequency was much lower than the 0.1 μm MF membrane. On the other hand, dosing 1 or 2 mg/L $FeCl_3$ had almost the same effect on flux decline. The normalized flux loss, over 60 min running time, decreased by 18% for both cases. Therefore, it is recommended to run the experiments with 1 mg/L $FeCl_3$ injection (see Fig. 8).

By looking at the plot of the 50 kDa UF membrane without using coagulant, the normalized flux dropped down and the loss was equivalent to 16.7% in flux

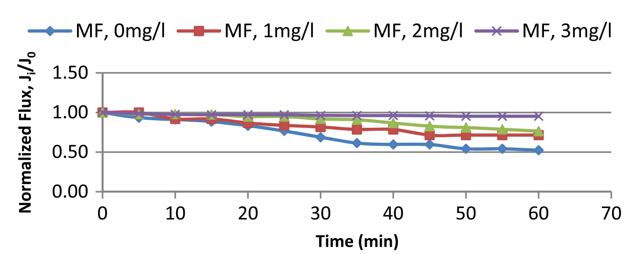

Fig. 7. Normalized flux vs. time, different coagulation doses, 0.1 μm MF membrane at constant pressure filtration mode.

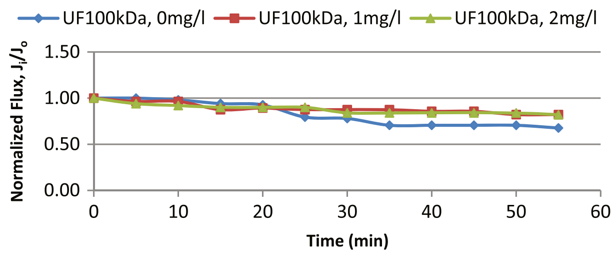

Fig. 8. Normalized flux vs. time, different coagulation doses, 100 KDa membrane at constant pressure filtration mode.

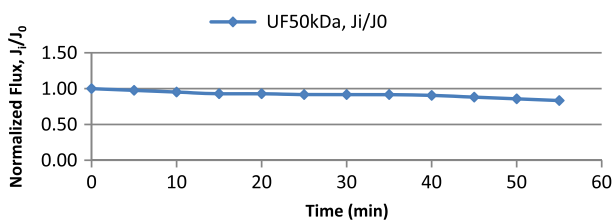

Fig. 9. Normalized flux vs. time, no coagulation, 50 KDa membrane at constant pressure filtration mode.

Table 6
Optimum $\Delta J_i/J_0$ of the three membranes used at constant pressure mode

Membrane	Configuration	$\Delta J_i/J_0$, %
MF 0.1 μm	1 mg/L $FeCl_3$	23.0
UF 100 kDa	1 mg/L $FeCl_3$	18
UF 50 kDa	No coagulant	16.7

rate. This ratio was better than the 100 kDa UF membrane case but the flux was less (see Fig. 9).

Optimum cases of constant pressure experiments for the three membranes are gathered in Table 6. It can be seen that the most preferable configuration is the UF 50 kDa membrane without coagulant followed by the 100 kDa UF membranes with 1 mg/L $FeCl_3$ and then the 0.1 μm MF with 1 mg/L $FeCl_3$ coagulant concentration because it has the highest $\Delta J_i/J_0$ reduction. However, it is important to notice that there is no big difference in the three $\Delta J_i/J_0$ reductions. By looking at another important factor which is the initial flux of each configuration, it is observed that the 0.1 μm MF with 1 mg/L has the highest flux (about the double of the 50 kDa UF membrane). Therefore, from an economical point of view the 0.1 μm MF membrane with 1 mg/L $FeCl_3$ injection is the most preferable option followed by the 100 kDa UF membrane with 1 mg/L $FeCl_3$ injection and leaving the least preferred option to the 50 kDa UF membrane without coagulation. Again, the last option (50 kDa) or the 100 kDa UF membrane without coagulation will become the best options in case of no possibility to use such chemicals due to strict environmental regulations, which is the case in many countries having big desalination market.

3.3. Permeate water quality analysis

The quality of permeate produced by the different membranes and configurations have been analyzed (Table 7). SDI values of all permeates were found less than 3 as recommended by RO membrane manufacturers. Values less than 2 were obtained for the 100 KDa and 0.1 μm membranes using coagulation. Similarly, very low turbidity values were obtained for all permeates with values as low as 0.06 for 50 KDa and 100 KDa UF membranes. Different values were found by several authors using different raw water quality. Kruger [32] found SDI values of less than one (SDI = 5 before UF) and turbidity less than 0.5 NTU when filtering Gulf water using UF membranes. Using dead end UF membranes to pretreat seawater, van Hoof et al. [33] found SDI values ranging from 0.5 to 3 and turbidity of less than 0.05. Our results confirmed that turbidity measurement was more consistent than SDI [34], where higher SDI values were found with smaller membranes pores sizes while MF/UF membranes work as barriers for defined particle sizes [35]. On the other hand, TOC removal was low due to lower size compared the used membrane pores. We noticed that the greater TOC removal has been achieved when using higher $FeCl_3$ dosage; i.e. 2 mg/L for 100 kDa and 3 mg/L for 0.1 μm membranes. Deeper investigation on NOM, biopolymers, humic substances, and transparent exopolymer particles (TEP) removal efficiency will be performed in the next phase of this study.

4. Conclusions

This study confirmed that the use of MF and UF membranes as pretreatment for SWRO systems has several advantages mainly providing very high and stable water quality to RO membranes. All the configurations using the three membranes with different pore sizes, with and without using coagulant, gave high quality of permeate with low SDI (2.1–2.88) and turbidity (0.06–0.18) values. However, the 0.1 μm MF membrane with 1 mg/L $FeCl_3$, followed by the 100 kDa UF membrane with 1 mg/L $FeCl_3$ injection, represent the most suitable and preferred options with an increase in 8 and 11.33% relative pressure, respectively, after one hour filtration.

Table 7
Water parameters analysis

	Seawater	UF 50 kDa w/o coag.	UF 100 kDa w/o coag.	UF 100 kDa +1 mg/L	UF 100 kDa +2 mg/L	MF 0.1 μm w/o coag.	MF 0.1 μm +1 mg/L	MF 0.1 μm +2 mg/L	MF 0.1 μm +3 mg/L
SDI (%/min)	11.91*	2.70	2.88	1.81	1.2	1.89	2.77	2.49	1.67
Turbidity (NTU)	0.60–0.75	0.06	0.14	0.06	0.06	0.17	0.18	0.10	0.13
pH	7.8–8.36	7.82	7.56	7.15	7.25	7.79	7.23	7.19	7.57
Temperature (°C)	20.2	20.4	22.6	21.3	21.3	24	24.5	25.5	22.5
TOC (mg/L)	1.94	1.652	1.82	1.76	1.05	1.665	1.481	1.62	1.06

*Value indicates SDI_5.
**Temperature taken while doing the analysis which is different than the temperature of water during the experiment (30 °C).

Without coagulation, the 50 kDa UF membrane had 19% of pressure increase by the end of the filtration period. At constant pressure filtration mode, the 0.1 μm MF membrane with 1 mg/L $FeCl_3$ injection, followed by the 100 kDa UF membrane with 1 mg/L $FeCl_3$, showed a decrease in 23.6 and 18% relative flux, respectively, after one hour filtration. However, 16.7% decrease in flux was observed for the 50 kDa UF membrane without using a coagulant.

From a techno-economical point of view (mainly product flow-rate, energy consumption, and cleaning frequency), the 0.1 μm MF membrane with 1 mg/L $FeCl_3$ has achieved the optimum case in this study. The second most preferable choice was the 100 kDa UF membrane with 1 mg/L $FeCl_3$. While the last preferred option, but still very efficient, is the 50 kDa UF membrane without coagulation. However, it should be kept in mind that not using a coagulant may not be a safe option to control membrane fouling and permeate water quality as raw water quality is susceptible to change during seasons and special events such as red tide, marine currents, or pollution. In addition, low concentration of coagulant could act as a safe option to control the process. In case of restrictions in using chemicals, membranes with smaller pores, i.e. the 50 or 100 kDa UF membranes without coagulation, would be the preferred options.

Deeper characterization and analysis of the organic, inorganic, and biological foulants responsible for flux decline as well as membrane fouling modeling and economic analysis of using MF/UF membranes in SWRO pretreatment replacing the conventional processes will be performed in the next phase of this study.

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