Brine Reject Dilution with treated Wastewater for Indirect Desalination

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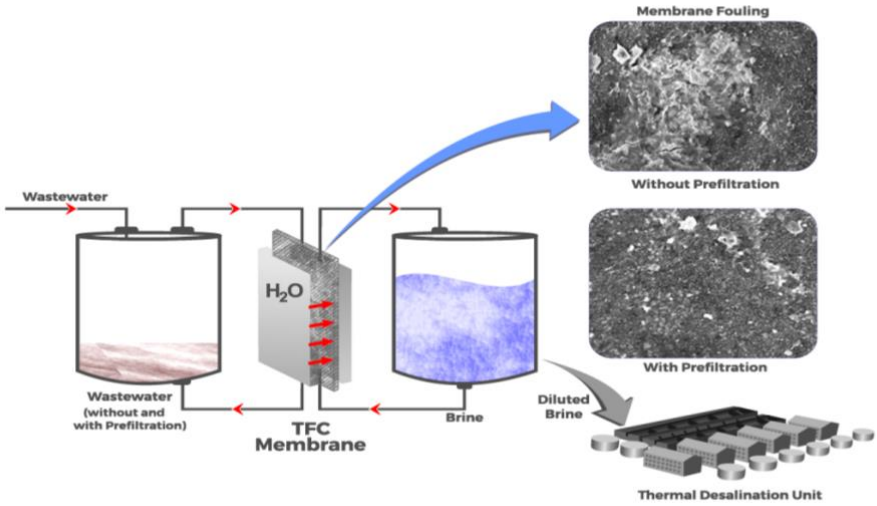
Graphical Abstract



Abstract

The forward osmosis (FO) process was suggested as a pretreatment to a multi-stage flashing (MSF) plant to reduce the environmental impact of brine discharge and the chemicals used. Yet, there is no study investigating the performance of the FO process pretreatment to the MSF plant using tertiary sewage effluent (TSE) as a feed solution. Combining MSF brine with the TSE generates a considerable permeation flux, reducing the membrane area and capital

cost. This study evaluated the performance of the FO process for indirect desalination of the MSF brine, considering membrane fouling, cleaning, required membrane area and the specific power consumption. The FO process used a thin-film composite (TFC) membrane to dilute the brine reject from the MSF plant by the TSE and hence converting waste solutions into a feasible water resource. A considerable high water flux (±35 L/m²h) was generated and slightly decreased throughout each experiment's 4 cycles. An enhancement in the water permeability was observed in the FO tests with a prefiltration of the brine reject and the wastewater with 20µm and an osmotic backwash cleaning of the used membrane. The prefiltration of the draw and feed solutions was effective in minimizing the impact of fouling. Maximum power consumption of 0.007 kWh/m³ was consumed in the forward osmosis process without prefiltration and decreased to 0.006 kWh/m³ in the FO process. The proposed FO system successfully diluted the brine reject' divalent ions, reducing their concentration to 43% in some cases. Depending on the FO membrane orientation, the TSE feed solution resulted in a 276% to 473% reduction in the number of FO elements required in the FO process compared to the seawater feed solution.

Keywords: Forward osmosis, Wastewater, Desalination, FO-MSF hybrid, Seawater

1. Introduction

Thermal desalination processes are broadly used to desalinate seawater in the Middle East (Mabrouk, 2013; Panagopoulos and Haralambous, 2020). MSF represents one of the main thermal desalination processes in the Gulf region (Buros, 2000). It has been proposed that maintaining the current MSF plants in better conditions for future use in the long term is less intricate than installing other desalination technologies (Mannan et al., 2019). Researchers in this domain has focused on tackling the in site issues of the MSF plants due to the high

efficiency of the process and its capability to treat elevated salinity feed solution (FS) (Thabit et al., 2019). One of the main issues affecting the MSF plant's performance is the non-alkaline scale fouling caused by calcium sulfate and magnesium sulfate precipitation on the plant's heat exchangers (El Din et al., 2002). Antiscalants, periodic cleaning and membrane technologies were suggested to treat the seawater and minimize the fouling in the thermal plants but cannot inhibit it (Mabrouk, 2013). The nanofiltration (NF) membrane process was proposed for seawater treatment, but experimental and pilot plant results revealed that it is not a cost-effective approach for seawater softening due to the high operation cost of the NF process (Mabrouk et al., 2015). Lately, the FO technique, a membrane separation process, was investigated to remove scaling ions from seawater (Altaee et al., 2014a; Thabit et al., 2019). In principle, the FO technique relies on the osmotic pressure gradient between the FS and the draw solution (DS) for freshwater extraction (Cath et al., 2006). The FO technique showed promising results such as low energy consumption, less fouling, high rejection rate and good water flux. In the FO-MSF system, MSF brine is diluted in the FO process and returns to the MSF for freshwater extraction and brine regeneration. The MSF top brine temperature (TBT) can be adjusted to control the plant's recovery rate for a continuous process. Despite the FO process advantages, it experiences intense concentration polarization (CP) during operation, reducing water flux. In addition, the relatively high osmotic pressure of seawater feed solution is another impediment controlling water flux in the FO process.

The hybrid system (FO-MSF) was proposed to remove undesirable multivalent ions from seawater using the MSF brine as a DS (Altaee et al., 2013). Combining the FO process with the MSF plant will reduce brine discharge to seawater since it will be the DS in the FO membrane for pure water extraction from the feed solution. Coupling the FO process with the MSF will

prevent the thermal pollution caused by brine discharge at 40 °C to seawater and the chemicals used, such as antiscalants and antifoaming, to control scales deposition onto the heat exchanger (Altaee et al., 2013). A wealth of literature discussing the environmental impact of improper treatment of the brine reject and proposes appropriate regulations to control brine discharge (Ariono et al. 2016). Alternatively, researchers proposed brine reject recycling using the FO process. Preliminary laboratory tests demonstrated the feasibility of applying this process for seawater pretreatment to the MSF plant (Hawari et al., 2018; Thabit et al., 2019). In laboratory-size experiments using MSF brine as a DS and seawater as an FS, the average water permeability in the FO test was 16.9 L/m^2h at 25 °C DS temperature (Thabit et al., 2019). When the temperature on the DS side was 40 °C, the average water flux reached 22.3 L/m^2h. Taking into consideration that 22.3L/m^2h water flux was high, a further enhancement in the water flux is always desirable for additional dilution of the DS. Unfortunately, the FO process suffers severe concentrative CP due to the high salinity of seawater feed solution. In the FO-MSF/MED system, the osmotic pressure between the brine reject and seawater is limited and cannot be increased due to the thermal plant's design conditions that operate at pre-designed top brine temperature (TBT). Recent studies revealed that the FO water flux increased by applying small pressure on the feed side in a pressure-assisted FO (PAFO) process (Khanafer et al., 2021). Using PAFO for brine reject dilution brought an insignificant increase in the specific power consumption due to the higher permeation flow. Nevertheless, higher water flux is still desirable to dilute further the brine reject and ensure scale prevention.

Earlier studies underlined the environmental impact of the brine reject from the MSF on the marine ecosystem when discharged to seawater (El-Ghonemy, 2018; Panagopoulos and

Haralambous, 2020). Brine reject contains chlorine, antiscalants, copper residues, and antifoaming agents of detrimental effects on flora and fauna when released to the seawater. The relatively high temperature of MSF brine results in thermal pollution when mixed with seawater. Some studies proposed the brine reject discharge into the sewer system. The latter approach would affect wastewater treatment because the high salinity of the brine reject inhibits the growth of the microorganisms. Instead of disposal as wastewater, tertiary sewerage effluent (TSE) could be used as an FS in the FO process to treat the thermal plant brine. Despite the severe freshwater shortage in the Middle East, large amounts of TSE are disposed of every day (Yangali-Quintanilla et al., 2011). The TSE salinity is about 1 to 2.6 g/L (Hawari et al., 2018), which is significantly less than the salinity of seawater in the Middle East (45 g/L), indicating its great potential for the dilution of the brine reject. TSE was previously used in the FO process for indirect desalination of seawater (Choi et al., 2017; Yangali-Quintanilla et al., 2011; Zhang et al., 2014; Zhao et al., 2012). Indirect desalination is an attractive concept in which wastewater streams, such as brine and TSE, are converted into a source for freshwater supply. Victor et al. used secondary wastewater effluent for the dilution of Red Seawater in the FO process to reduce the cost of seawater desalination. Coupling wastewater effluent with the Red Sea water resulted in a 50% reduction in the specific power consumption for desalination by 50%, around 1.5 kWh/m^3. The study reported a 28.8% water flux decline after 10 days due to the FO membrane's fouling. However, cleaning with air scouring and freshwater could recover 98.8% of the initial water flux. Another study by Choi et al. studied wastewater reclamation by the FO process using a NaCl (0.6 M) DS to resemble the concentration of the seawater (Choi et al., 2017). The study used a special thin-film composite (TFC) membrane with functionalized carbon nanotubes (CTN). Results indicated that water flux was lower in the fabricated membrane than that in commercial TFC

membranes. Experimental results also showed the great potential of coupling seawater with treated wastewater effluent for indirect seawater desalination. However, there is no experimental study on treated wastewater application for the dilution of thermal plant brine by the FO process.

The present study is the first experimental work on applying the FO technique for the treatment of brine reject from the MSF plant using treated wastewater as FS (**Fig. 1a)**. There are several advantages of using brine reject and treated wastewater in the FO process: reducing wastewaters disposal to seawater, converting wastewaters into a viable source of freshwater, and minimizing scale fouling of the MSF thermal plant. Coupling low-salinity wastewater with brine reject at 40 °C would enhance water flux in the FO process and reduces the membrane required in the pretreatment process. The specific power consumption and the membrane elements required in the FO process using a TSE feed solution was compared with that of a seawater feed solution. The targeted dilution percentage of the brine reject by the FO process is 14% or higher, corresponding to the recovery rate in the MSF plant operating at a performance ratio equal to 8 (Altaee and Zaragoza, 2014; El-Ghonemy, 2018; Morin, 1993). It also aimed to provide insights into the development of lower energy and cost-saving desalination hybrid systems.

2. Materials and experiments

2.1 Stream solutions

In this study, treated wastewater and brine reject were the feed and DS of the FO process, respectively. Wastewater samples were provided by the Blacktown wastewater treatment plant, Sydney, Australia. **Table 1** presents the average (triplicate values) concentrations of the

wastewater and the seawater brine. To prepare the DS, seawater was collected from Sydney, New South Wales, and heated to reach the brine reject concentration of the MSF thermal plants in the Middle East, circa 80 g/L (Hawari et al., 2018). The DS's temperature was maintained at 40 °C during the experiments, with 25 °C at the FS's temperature. The DS (brine reject) osmotic pressure is about 70.4 bar, and the FS (wastewater) is about 0.804 bar. A microfilter of 20 µm size was used for removing the turbidity and particulate organic matters from seawater and wastewater in selected FO experiments to study the pretreatment of feed and DS impact on the process performance.

Table 1: (a) Characteristics of wastewater and brine. (b) Comparison of wastewater and brine turbidity and total organic carbon (TOC) before and after microfiltration.

a	Parameter	Wastewater	Brine Reject
	TDS (g/L)	0.967	80.2 ± 0.5
	Conductivity (mS/cm)	3.91 ± 0.27.5	106.7 ± 1.2
	pH	7.2 ± 0.2	8.35 ± 0.5
	Ca^{2+}(ppm)	-	1039 ± 21
	Mg^{2+}(ppm)	-	2200 ± 34
	SO_4^{2-}(ppm)	-	6566 ± 43
	K^+ (ppm)	-	873 ± 19
	Na^+(ppm)	-	19152 ± 82
	Cl^- (ppm)	-	22352 ± 78

b	Solution	Turbidity (NTU)	TOC (mg/L)
	Wastewater (no prefiltration)	7.1 ± 0.1	49.69 ± 2
	Wastewater (with prefiltration)	1.76 ± 0.12	28 ± 1.3
	Seawater (no prefiltration)	2.43 ± 0.15	30.6 ± 0.5
	Seawater (with prefiltration)	0.9 ± 0.05	9.4 ± 0.2

2.2 Membrane and equipment

The membrane implemented in the experiments in this study was the TFC FO membrane manufactured by Porifera. The selection of this membrane was based on its potential to achieve high water flux which is usually attributed to the thin support layer (SL) structure of the TFC membranes that reduce the internal concentration polarization (ICP) (Wang et al., 2015). 344 microns is the structural parameter of the TFC membrane. The latter tolerates feed temperature of 40 °C and reject salt up to 90%. **Table 2** summarised the membrane's chemical and physical characteristics.

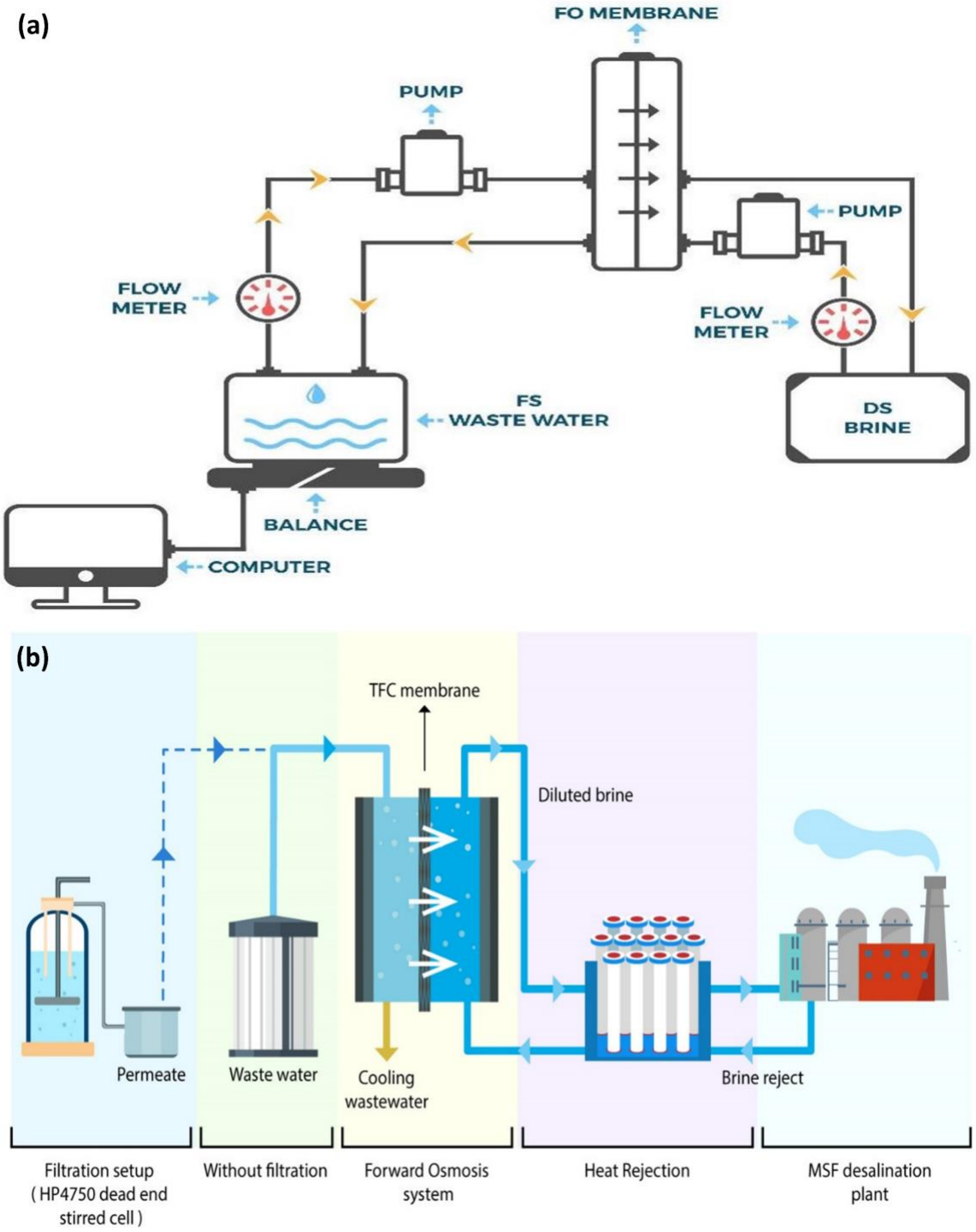
Table 2: TFC membrane: physical and chemical properties.

Parameters	Value
Membrane used	TFC Porifera
Material	Polyamide
A_w(L/m^{2}h.bar)	2.1
B(kg/m^{2}h)	1.2
Active layer water contact angle (°)	68.5 ±0.7
Support layer water contact angle (°)	53.9 ± 2
Zeta potential (mV)	-41.9 ± 2.44
pH tolerance	2-11

Note. A_w: the coefficient of membrane permeability & B: the salt permeability coefficient.

Fig. 1a shows the experimental design of the FO process used in this study. The FO membrane of 42 cm² was placed in the CF042A-FO cell of 5×4×3.25-inch exterior dimensions (Sterlitech, USA). The membrane was positioned in two orientations depending on the experiment, either with the active layer (AL) faces the FS (AL-FS), also called the FO mode or in the PRO mode in

160 which the SL faces the FS (AL-DS). Blue-White Industries Ltd F-550 flow meters were used to
161 monitor the flow rates, and two pumps manufactured by Cole-Parmer maintained the
162 circulations of the FS and DS. The conductivity of solutions, the salinity and the TDS, were
163 measured using HACH, HQ.14d portable conductivity meter. The turbidity of the solutions
164 was measured using a turbidity meter, HACH 2100P. Further, **Fig. 1b** illustrates the propsed
165 FO-MSF hybrid system.



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167 **Fig. 1a.** Lab-scale experimental design of the FO system. Operational parameters: 2 LPM flow
168 rate, 25 °C and 40 °C on the feed and draw sides, respectively. (b) An illustration of the
169 proposed FO-MSF hybrid system.

170 The wastewater weight variation (ΔW) was included in the following equation to calculate
171 the water flux:

$$J_W = \frac{\Delta W}{A.\Delta t} \quad \text{(Eq. 1)}$$

ΔW: wastewater weight variation (kg), A: area f the FO membrane (m^2) and Δt: time (h).

The filtration was conducted using HP4750 dead-end stirred cell. A 20 micron Whatman membrane sheet was cut and placed against the porous support disc and sit perfectly without bending or extending outside to avoid leakage. The diameter of the flat sheet is 47-48 mm, with an active membrane area of 17.3 cm^2. All of the experiments were carried at a constant pressure of 1 bar and 20 °C.

2.3 Experimental work

Brine reject solution was prepared in the laboratory by concentrating seawater on the MSF brine's concentration level in the Middle East. Each FO experiment was operated for 180 min, membrane washed for 30 min and then repeated 3 times to make 4 cycles in total for each experiment. The volume of wastewater and brine reject was 5 L at the start of each experiment, while the temperature was 25 °C and 40 °C, respectively. A constant flow rate of 2 LPM was used in the FO cell, using the Porifera TFC FO membrane. Each experiment started with a new TFC membrane, the latter was soaked in DI water for 30 min before the first cycle, and at the end of each cycle, the membrane was cleaned and reused for the other 3 cycles. In **Table S1**, a summary of the experimental work is provided; each experiment was run 4 times using the same membrane following the same cleaning method. In all experiments, one variable was changed at a time, and its impact on the FO performance was investigated. In the FO filtration experiments, the FS and DS prefiltration and membrane orientation were the variables to investigate their impact on water flux, ions rejection, reverse salt flux, specific power consumption, and membrane fouling. In Exp1, AL-DS was the membrane orientation,

and the cleaning of both layers was at a flow rate of 2 LPM with 40 °C DI water for 30 min. Experiments 1, 3, 5 &7 were run in the AL-DS orientation and Exp2, 4, 6 & 8 in the AL-FS mode. Exp1-4 were performed without the prefiltration of feed and DSs while Exp5-8 with filtration. In Exp2 and 4, membranes were cleaned with 40 °C DI water and 3 LPM flow rate. The prefiltration of the DS and the FS was done in Exp5 and 7, and both membranes were washed with DI water at 3 LPM flow rate. Similarly, for Exp6 and Exp8, filtered streams were used in the FO experiment. In the cleaning process, the flow rate of the cleaning solution and osmotic backwash were varied to evaluate their impact on water flux recovery. The membrane backwashing was performed with 40 g/L NaCl solution at 40 ^{o}C and 3 LPM flow rate on the SL of the membranes and DI water at 40 °C and 3 LPM flow rate on the AL. The strategy of backwashing with NaCl showed promising results as it helps remove foulants from the membrane's SL pores (Yu et al., 2017). In the MSF desalination plant, 40 °C temperature could be obtained from the brine reject using a heat exchanger.

2.4 Analytical processes

The experimental plan was designed to inspect the effect of key parameters such as the membrane orientation, the reversible fouling, and the membrane cleaning strategies on the FO process. Eight experiments of 4 cycles were carried out under several operational parameters. The membrane cleaning methods, the orientation and the treatment of the brine reject, and the wastewater were the changing parameters

To examine the performance of the FO process under defined experimental conditions, the water flux was calculated for each run using the recorded weight change of the wastewater. Flux reduction (FR) was calculated after each cleaning method to study the effectiveness of

these methods in water flux recovery. The latter was calculated by applying the following expression:

$$FR = \left(1 - \frac{J_c}{J_f}\right) \times 100 \qquad \text{(Eq. 2)}$$

J_c: is average water flux after cleaning and $J_{f:}$ average water flux before cleaning.

The reverse solute flux (RSF) was calculated to understand the behavior of the FO membrane. RSF is the solute penetration in the membrane from the DS side towards the FS side and is a consequence of the difference in the solute concentrations. The values of RSF ($J_{S,}$ g/m²h) were studied and analyzed using the equation:

$$J_S = \frac{(Ct.Vt - C0.V0)}{A.\Delta t} \qquad \text{(Eq. 3)}$$

In equation 3, C0 and Ct in (g/L): solute concentrations at the beginning and at time t, respectively. V0 and Vt in (L): volumes of the FS measured at the beginning and at time t, respectively, A in (m²): the membrane area, and Δt in (h): the allocated time.

To investigate the impact of the FO technique conducted in the experiments, it is important to calculate the energy consumption (E, kW h/m^2) in the standalone FO process as per the following:

$$E = \frac{P_f Q_f + P_D Q_D}{n Q_p} \qquad \text{(Eq. 4)}$$

The energy consumed in the prefiltration method was calculated using the expression below:

$$E = \frac{P_f Q_f}{n Q_p} \qquad \text{(Eq. 5)}$$

P_f: wastewater hydraulic pressure in bar, Q_f: FS flow rate in m^3/h, P_D: brine hydraulic pressure in bar, Q_D: DS flow rate in m^3/h, n= 0.8: the pump efficiency, Q_p: permeation flow in m^3/h.

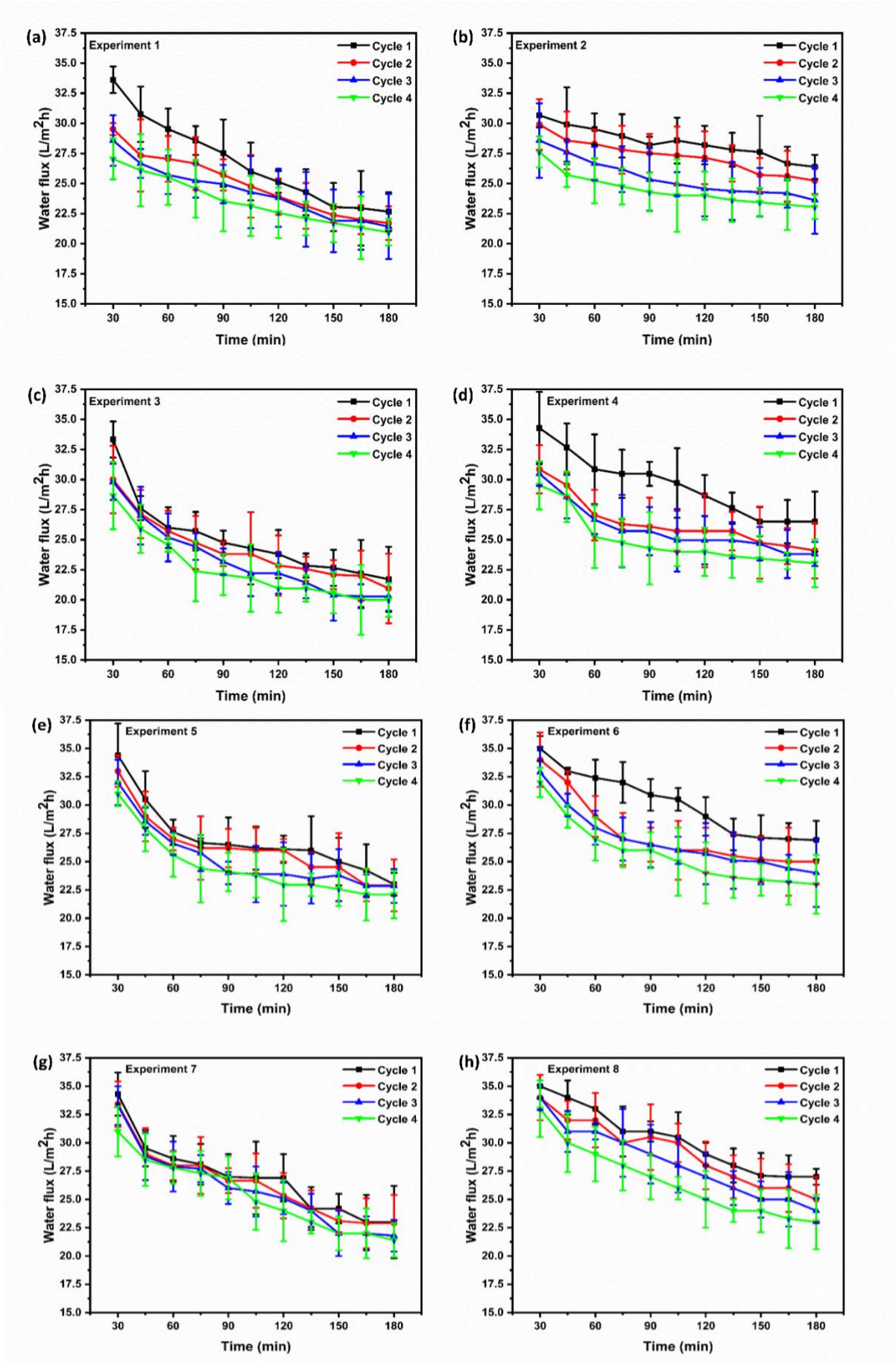
3. Results and discussions

3.1 Water flux in the FO process

The water flux was calculated every 15 min in each cycle of 180 min, using Eq. 1, and the results are presented in **Fig. 2.** The latter showed the variation of water flux over the operational time without the prefiltration of the FS, and DS. **Fig. 2** illustrated the water flux decline that is explained due to the decrease in the pressure difference across the membrane as a result of the water permeation and membrane fouling. As such, the driving force reduced, causing a reduction in the water flow (Zhang et al., 2014). As shown in **Fig. 2a**, water flux in Exp1 (AL-DS) was 33.6 LMH in cycle 1 but decreased 32%, reaching 22.7 LMH after 180 min. The performance of the FO system was studied after cleaning the fouled membrane with DI water at 40 ^oC and 2 LPM flow rate. The reason for using DI water at 40 ^oC for membrane cleaning is its ability to dissolve and remove the fouling materials (Ibrar et al., 2020). Comparing to the initial water flux in cycle 1, it decreased to 29.5 LMH in cycle 2. The initial water flux at the beginning of cycles 3 and 4 was 28.6 and 27.0 LMH, respectively. The slight decline in the initial water flux in cycles 2 to 4 compared to cycle 1 is due to the irreversible membrane fouling that was not removed by cleaning with hot DI water. After cleaning with 40 ^oC DI water at a 2 LPM flow rate, the initial water flux in cycle 4 was 27.0 LMH, and it declined to 21 LMH at the end of cycle 4 of Exp1. Despite cleaning with 40 ^oC DI water at a 2 LPM flow rate, the results show 22% decline in permeation flux at the end of cycle 4 of Exp1. To study the impact of the membrane orientation, Exp2 was conducted in the AL-FS mode. For the same cleaning method (**Fig. 2.b**), water flux in the first cycle of Exp2 was 30.7 LMH

and declined by 14% to 26.4 LMH at the end of cycle 1. At the beginning of cycles 2, 3, and 4 the initial water flux was 29.9, 28.6, and 27.6 LMH, respectively, with 17 % water flux decrease by the end of cycle 4. At this point, the drop in the water flux under the AL-DS mode was quicker, which might be explained due to the combination of dilution and more intense membrane fouling. This observation could be due to the active and SL structure, leading to more severe concentration polarization in the FO mode, which has been investigated in previous works (Ibrar et al., 2020; Vu et al., 2018).

Exp3 and Exp4 studied the effect of the flow rate of DI water in the cleaning cycle on the FO process in AL-DS & AL-FS modes. The 40 ^{o}C DI water flow rate in the cleaning process was increased to 3 LPM in Exp3 and Exp4. As presented in **Fig. 2.c** and **Fig. 2.d**, the initial water flux of cycle 2 was 30.0 and 30.9 LMH, respectively, higher than the initial water flux of cycle 2 in Exp1 and Exp2, **Fig. 2.a** and **Fig. 2.b**. This suggests that cleaning at 3 LPM flow rate has a better outcome than at 2 LPM as it was more efficient in removing fouling materials from the membrane surface. Therefore, DI water at 3 LPM was used for the membrane cleaning for the rest of the FO experiments.


(a)
Experiment 1
(b)
Experiment 2
(c)
Experiment 3
(d)
Experiment 4
(e)
Experiment 5
(f)
Experiment 6
(g)
Experiment 7
(h)
Experiment 8
Cycle 1
Cycle 2
Cycle 3
Cycle 4
Water flux (L/m2h)
Time (min)

272

Fig. 2. The water flux readings in the FO process, (a-d) without FS and DS prefiltration and (e-h) with FS & DS prefiltration, were conducted in both membrane orientations following 3 cleaning runs.

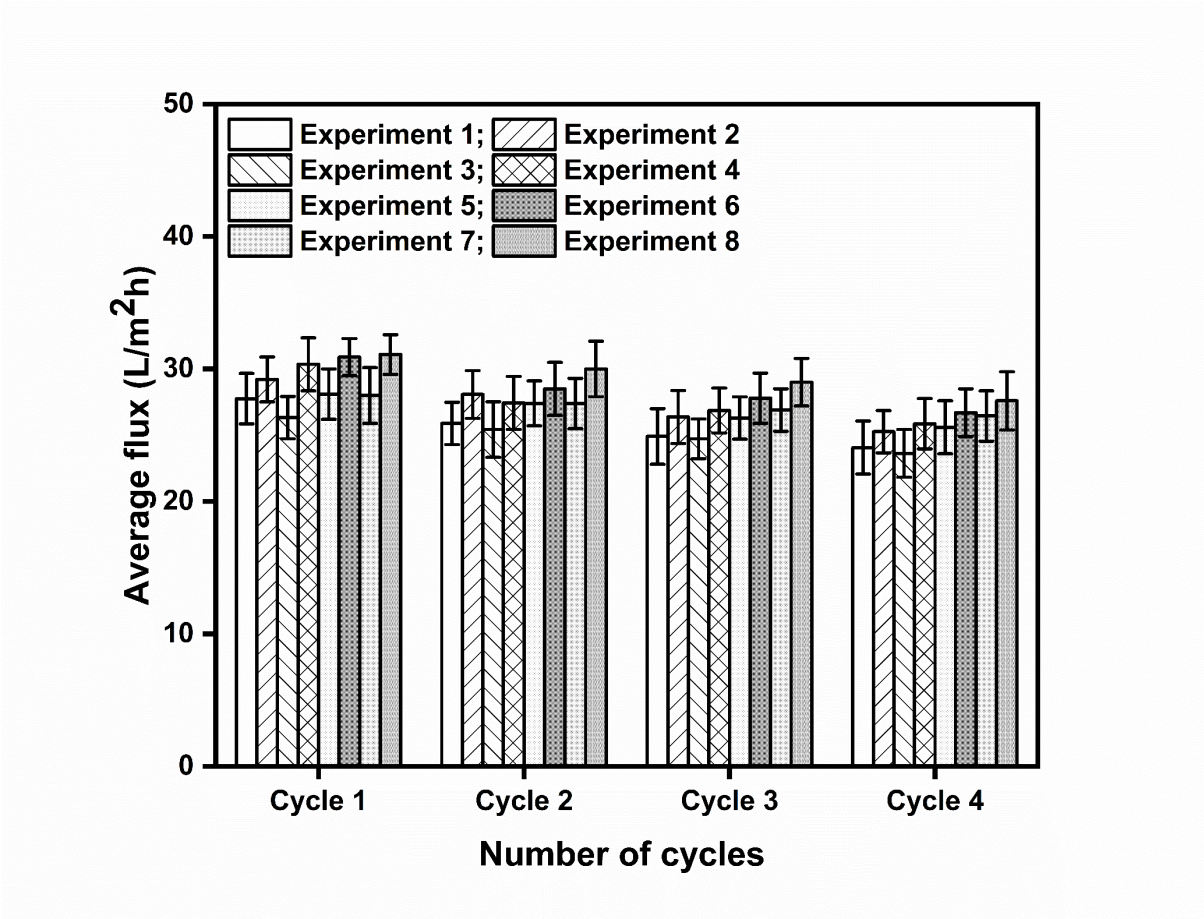
3.2 Impact of prefiltration and osmotic backwash

Since the flow rate of 3 LPM in the previous experiments has shown better performance, experiments 5 to 8 were performed with 3 LPM cleaning flow rate. The stream solutions in Exp5-8 were prefiltered using 20 µm Whatman filter to reduce the turbidity and the TOC of both wastewater and the brine reject solution (**Table 1b**). Water flux in cycle 1 was 34.4 LMH in Exp5 and 35 LMH in Exp6, as presented in **Fig. 2.e and Fig. 2.f,** respectively**.** After membrane cleaning with 40 °C DI water, the initial water flux was 33.0, 32.0, and 31.0 LMH in cycles 2, 3, and 4 of Exp5 and 34.0, 32.9, and 32.0 LMH in cycles 2, 3, and 4, respectively of Exp6. Water flux in Exp5 and Exp6 was higher than Exp3 and Exp4, which was carried out without the prefiltration. This improvement is correlated with reducing the turbidity and the TOC of both FS and DS. The micro size compounds that usually accumulate on the FO membrane were eliminated with the prefiltration. This finding agrees with a previous experimental work that achieved higher water flux in the FO membrane after the feed solution prefiltration (Hawari et al. 2018).

The study also investigated the impact of introducing the osmotic backwash of the membrane SL on the cleaning strategy at the end of each cycle. Exp7 showed the results of the osmotic backwash in which the SL was cleaned with 40 °C NaCl 45g/L at 3 LPM flow rate and the AL with 40 °C DI water at 3 LPM flow rate. This cleaning method removes fouling materials accumulated on the membrane layers by combining osmotic backwash and cross-flow cleaning with hot water. Results revealed a maximum water flux of 33.42 LMH in cycle 2 that

dropped to 23 LMH at the end of cycle 3 (**Fig. 2.g**)**.** Moreover, when comparing the same cleaning method in the AL-FS orientation in Exp8, it is shown that the initial water flux was 35.0, 34.0, 34.0 and 33.0 LMH in all the 4 cycles respectively and declined steadily to reach 27, 25, 24 and 23.0 LHM at the end of each cycle (**Fig. 2.h**). All the water flux values measured in the TSE experiments were considered high compared with the water flux of the FO process conducted with seawater FS(Khanafer et al., 2021). The maximum water flux recorded using TFC membrane with brine reject as DS and seawater as FS was 7.4 LMH. The results highlight the effectiveness of applying the FO process for wastewater and brine recycling to maintain a considerably high water flux and minimize the environmental impact of wastewater disposal.

According to **Fig. 3,** the average flux decreased in the consecutive cycles in all experiments. For example, the average flux was 27.8 L/m^2h in the first cycle of Exp1 and decreased to 24.1 L/m^2h by the end of cycle 4. The maximum average permeation flux of 31.1 L/m^2h was recorded in Exp8 (AL-FS), and the membrane was cleaned using an osmotic backwash. Overall, the Al-FS tests yielded a better average permeation flux than the AL-DS tests.



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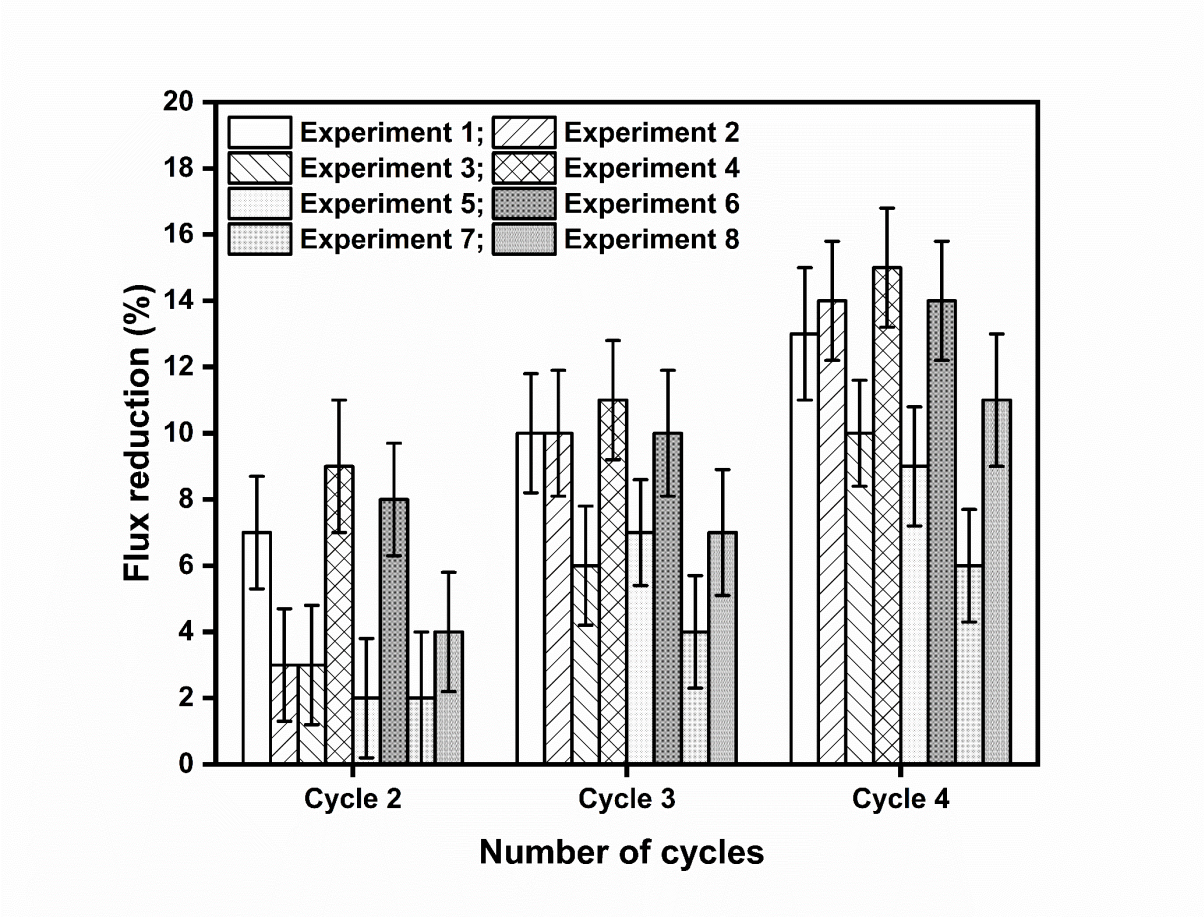
312 **Fig. 3.** Average flux for each cycle in the FO experiments. Each FO process was run for 4
313 consecutive cycles. Each FO cycle was operated for 180 min, and the water flux was measured
314 every 15 min. Experiment (1-4) without FS & DS filtration and (5-8) with FS & DS filtration.

315 **3.3 Flux reduction**

316 The cleaning of the used and fouled membrane was either with 40 ^{o}C DI water or backwashed
317 with 40 ^{o}C DI water and NaCl solution. The reduction in water flux was calculated using Eq. 2,
318 and results are available in **Fig. 4**. Water flux reduction was generally lower in the AL-DS than
319 in the AL-FS tests due to the greater membrane fouling in the latter test. For example, water
320 flux reduction at the end of cycle 4 of Exp1 and Exp2 was 13% and 14%, respectively, indicating
321 that water flux reduction was higher in Exp2 when the FS faces the AL of the membrane.
322 Experiments also revealed that a better cleaning process was achieved at 3LPM flow than

2LPM flow rate, especially when the DS is against the AL of the membrane. Water flux decrease at the end of Exp3 was 10% compared to 13% at the end of Exp1 (**Fig. 4**). The higher flux reduction in the AL-FS could be due to the high hydrophilicity of the membrane AL (**Table 2**), which promoted the organic fouling. Fouling experiments showed that membrane fouling was more severe when the FS (wastewater) faces the membrane SL (Appendix A).

The prefiltration of FS and DS improved the filtration process and reduced water flux reduction in the consecutive filtration cycles 1 to 4. For example, the water flux decline at the end of Exp5 was 9% compared to 10% at the end of Exp3, performed at the same operating and cleaning condition but without filtration. It should be pointed out that the prefiltration of FS and DS was less efficient in preventing fouling in Exp6 (AL-FS) due to the ineffectiveness of the cleaning process. Combining osmotic backwash with DI water cleaning at 40 °C resulted in better cleaning and water flux recovery. The osmotic backwash improved removing fouling materials trapped in the membrane SL while cleaning the AL with 40 ^{o}C DI water at 3LPM flow rate was considered effective for cleaning the AL in Exp7 recorded 6% water flux reduction after 4 cycles. Therefore, the best operating and cleaning methods are presented in Exp7, in which 40 °C DI water and osmotic backwash at 3 LPM flow rate are used for the membrane cleaning.



340

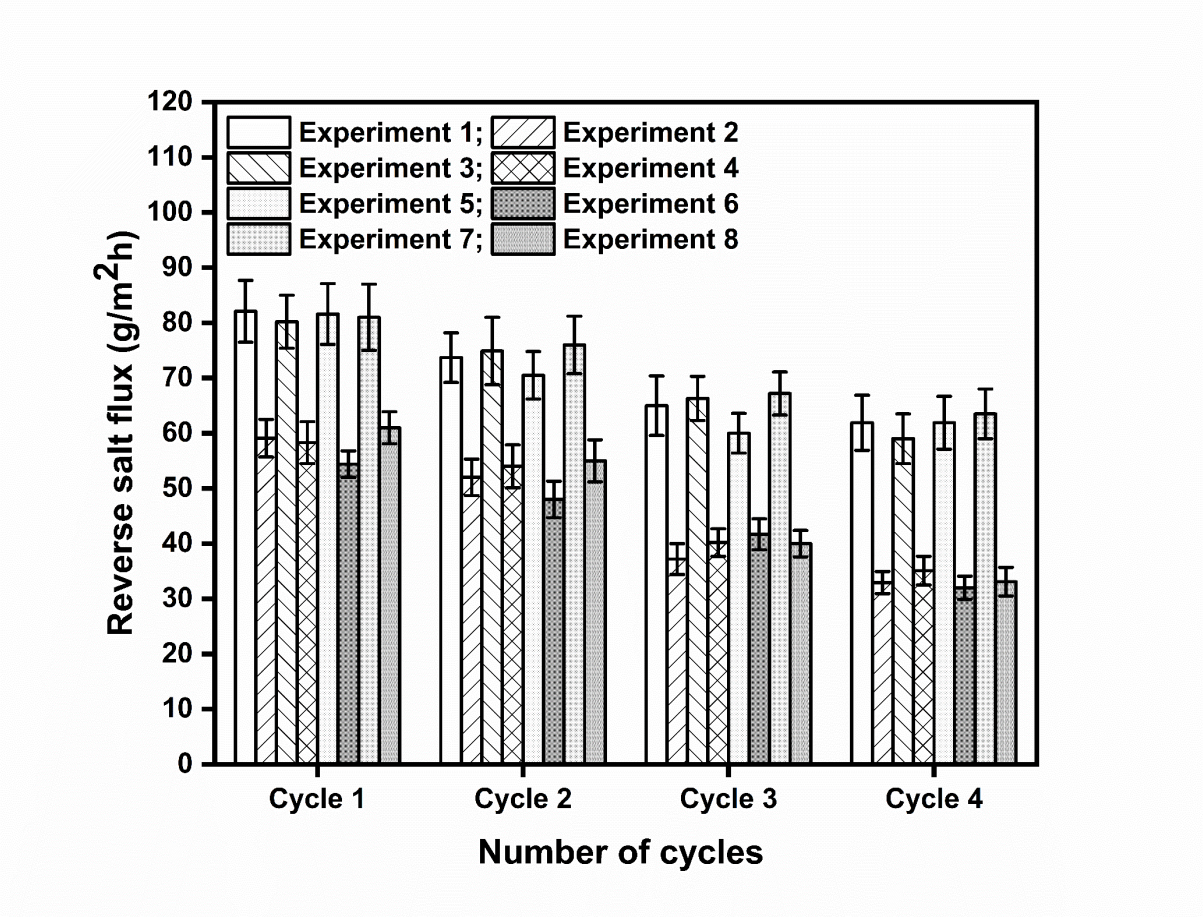
341 **Fig. 4.** Flux reduction in the FO experiments after each cleaning method. Membrane cleaning
342 was performed after each cycle for both AL-FS and AL-DS orientations. No FS & DS
343 prefiltration in Experiments (1-4) and with prefiltration in (5-8)

344 **3.4 Reverse solute flux**

345 Reverse Solute Flux is a major challenge that needs investigation in the FO process due to the
346 diffusion of salt leakage towards the FS side. RSF would decrease the concentration gradient,
347 lose draw solute, and increase fouling (Oh et al. 2014). Controlling and reducing RSF was
348 studied, and literature showed interest in operational strategies and membrane development
349 that can collaborate in minimizing the reverse diffusion of draw solutes toward the FS (Zou et
350 al., 2019). The reverse salt permeation is strongly related to the TDS of the DS. In this study,
351 the TDS of the wastewater and the seawater were, respectively, 0.963 and 80.2 g/L. The RSF

(J_S, g/m^{2}h) was calculated following Eq. 3, and the results are presented in **Fig. 5**. The latter shows the RSF in each cycle following different cleaning strategies. The RSF in the AL-DS for cycle 1 was 82.1 g/m^{2}h, and 80.2 g/m^{2}h for Exp1 and Exp3, respectively and 81 g/m^{2}h of Exp5 & Exp7 and decreased to 61.9, 59.0, 61.0, and 63.5 g/m^{2}h in cycle 4 of Exp1, Exp3, Exp5 and Exp7, respectively. Whereas for the FO experiments in the AL-FS mode, 59.1, 58.1, 54.4, and 61.0 g/m^{2}h were the RSF in cycle 1 of Exp2, Exp4, Exp6, and Exp8 reached 32.9, 35.1, 32.0, and 33.1 at the end of cycle 4. As for the orientation of the FO membrane, results revealed that the RSF is higher in the AL-DS mode. This can be explained due to severe concentration polarization (CP) when the DS faces the membrane's SL, leading to a reduced concentration of the DS inside this layer; hence, the RSF was lower in the AL-FS tests. Permeation flux dilutes the draw solute, creating a dilutive internal CP, which, in turn, reduces the concentration of the DS in the SL. This phenomenon is less severe in the AL-DS mode as the DS faces the AL (Oh et al., 2014).

It is also shown in **Fig. 5** that almost all the values of the reverse flux decreased after each cycle, i.e., cycle 1>cycle2>cycle3>cycle4. The decline in the RSF in each cycle after cleaning is due to the irreversible fouling of the membrane, which reduced the water flux and the RSF simultaneously in the FO process. In general, RSF is less in the FO processes performed in the AL-FS mode or when the FO membrane is fouled. In the current application, both feed and DSs are waste streams, and hence contamination of the FS due to RSF is not a concerning problem. However, the diffusion of salts from the draw side could slightly reduce the driving osmotic force of the process. Nevertheless, wastewater FS of about 1 g/L initial TDS and brine reject DS from the MSF plant provides a significant osmotic pressure for the FO process.



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376 **Fig. 5.** Variation of RSF without FS & DS prefiltration in Experiments (1-4) and with prefiltration
377 in Experiments (5-8). Each experiment consists of 4 cycles of FO processes. The membrane
378 was cleaned after each cycle.

3.5 Tackling fouling materials

The deposition of wastewater materials on the active and SL and within the porous SL reduces water transportation and affects the overall membrane performance. Fouling might result from ions scaling, the accumulation of organic or colloidal materials and microbial growth (Ibrar et al., 2019). External fouling occurs on the membrane AL in the AL-FS orientation; however, internal fouling is more intense where foulants of a smaller size than the membrane pores penetrate the SL, leading in some cases to pores clogging. This type of fouling is popular when the FS faces the SL (Zhao et al., 2016). In this study, physical cleaning strategies were implemented to mitigate the fouling materials, and a combination of analysis techniques was conducted on the tested membranes. Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Spectroscopy (EDS) was used to identify the elemental composition of the fouling materials, and Fourier Transform Infrared Spectroscopy (FTIR) in its turn is best for identifying the presence of organic materials.

3.5.1 SEM/EDS Scanning of fouled membranes

It is important to mention that the fouling matters are not evenly located on the membrane surface (Ping Chu and Li, 2005). The scale morphology varies within the same membrane and with other membranes, illustrated in **Fig. 6**. The deposited foulants mostly covered the membrane surface with various structures and shapes that might indicate a combination of different fouling compounds. The prefiltration of the stream solutions reduced the precipitated fouling compounds that were not highly presented in the SEM images compared to the images of the membranes when FO processes were conducted without prefiltration. The fouling compounds on the AL in the FO mode without prefiltration of the stream solutions

were bigger and different in morphology. The filtration for the same membrane orientation minimizes the presence of a larger accumulation of foulants.

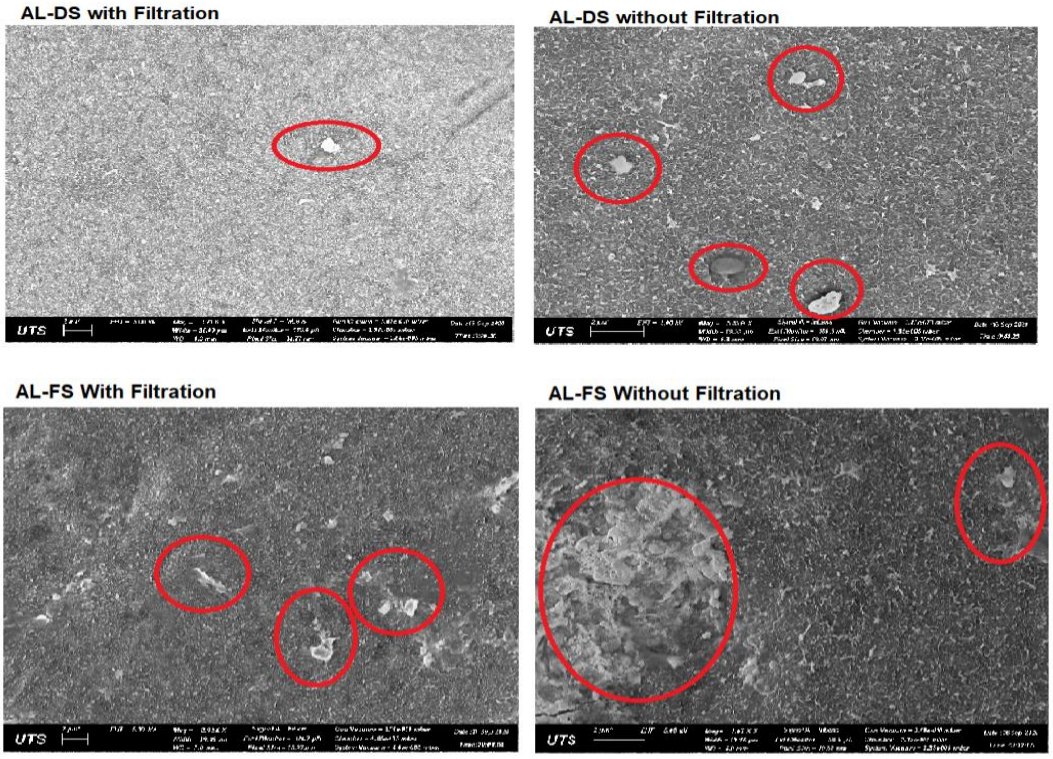

Fig. 6. SEM images of the AL of the fouled FO membranes. Fouling on the AL are more remarkable without filtration and in AL-FS according to the membrane samples used.

SEM and EDS scanning were coupled to visualize the fouling compounds distinctly, characterize the fouling layer and obtain information on its chemical composition. According to the EDS spectrums, the organic compound (C &O) and other inorganic ions such as Mg, Ca, and Fe mainly showed spectra **(Fig. S1** & **Table S2)**. This is in agreement with the SEM-EDS images where fouling particles were displayed and identified in colors (**Fig. 7**).

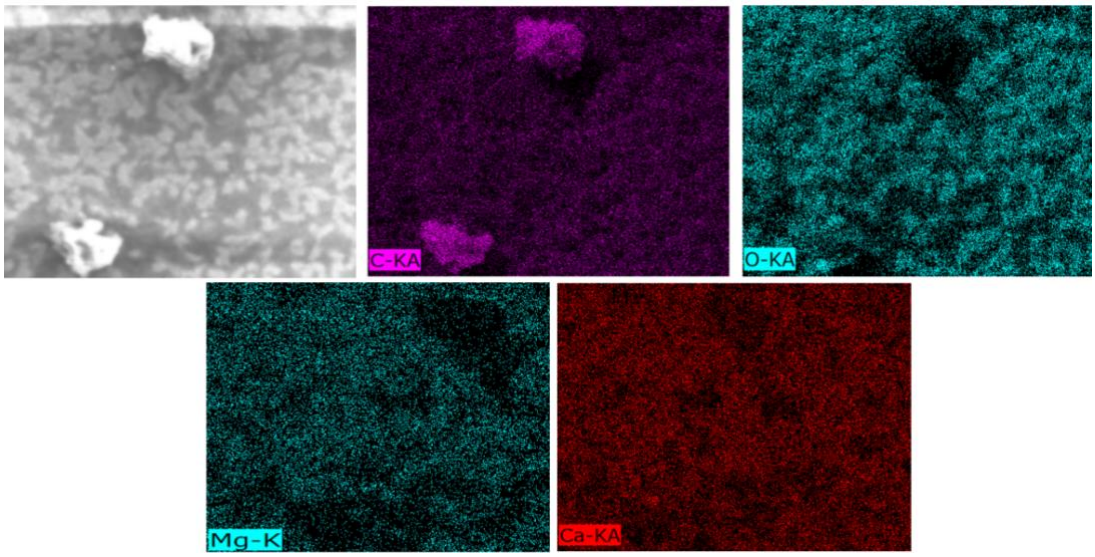

Fig. 7. SEM-EDS analysis of the AL of the fouled FO membrane in the AL-DS orientation at the end of cycle 4 after hot DI water cleaning. C, O, Mg and Ca are clearly observed on the surface of the sample membrane.

3.5.2 FTIR Analysis

FTIR technique has been applied widely to identify the functional groups of the foulants on the membrane by attenuated total reflection (ATR). In other words, inorganic and organic fouling compounds that absorb the radiation-specific compound can be characterized by FTIR spectra. Both fouled and washed membrane layers were scanned using an FTIR scanning microscope and later compared to a new membrane's spectra. A clear change is observed in the FTIR spectrum of the used membranes in both orientations compared to the pristine ones. The spectroscopy also shows that the spectrum of the membranes after cleaning resemble the new membranes, which can explain the efficiency of the cleaning strategy. On the used membrane (AL-FS) and the FTIR of the AL, the two peaks observed at a wavenumber of 875 cm^{-1} and 1875 cm^{-1} differentiate the fouled membrane from the new and washed membranes. In addition, the diminishing of a few peaks in the bands at wavenumber 500-850 cm^{-1} and 1000-1300 cm^{-1} is noticeable in the spectrum (**Fig. 8a**). Similarly, the FTIR on the SL

indicates the presence of fouling materials and how the cleaning methods enhanced the performance of the fouled membrane (**Fig. 8b**).

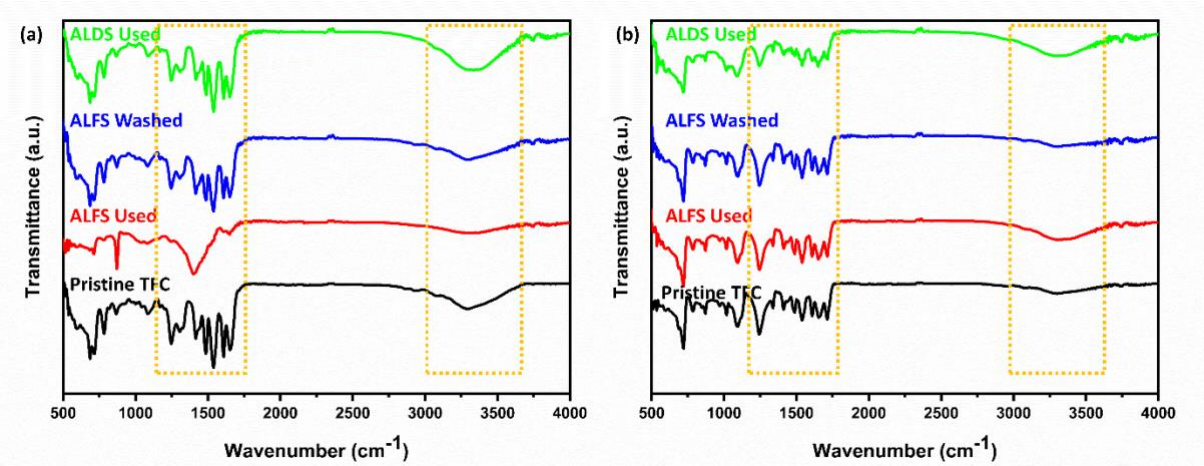

Fig. 8. FTIR spectroscopy of the new, fouled and washed membranes. (a) Scanning the AL in the AL-FS mode. (b)Scanning the SL in the AL-DS mode.

3.6 Energy consumption and membrane cost

The main component in the cost of desalination is the energy requirements; therefore, this section focuses on the energy consumed by the FO system during the ongoing process. The energy consumption was calculated using Eq. 4, and the results were illustrated in **Fig. 9.** The energy consumed in the prefiltration process of the solutions was calculated using Equation 5 and 0.034 kW h/m^2 was the amount of energy consumed for the prefiltration before the FO process. This extra energy is not added to the values presented in **Fig. 9**. Results revealed that the specific power consumption increased from cycle 1 to cycle 4. This might be due to the lower permeation flow in cycle 4 that caused an increase in the specific power consumption, as explained in Eq. 4. Increasing the flow rate from 2 to 3 LPM did not increase the power consumption in Exp3&4 (0.006 & 0.005 kW h/m^2); the explanation for this would be due to the higher water flux was achieved after cleaning with 40°C DI water at 3LPM. For the same reason mentioned above, results revealed that the DS and the FS filtration decreased the

448 energy consumption of the standalone FO process compared to the FO experiments without
449 the filtration of the FS and DS. The FO process energy in this study is considered very efficient
450 compared with the nanofiltration pretreatment process (Altaee et al., 2013). The FO process
451 maximum energy consumption in this study was 0.007 kW h/m^2, which is considered very low
452 compared to the energy consumption when seawater was used as FS. 0.020 kW h/m^2 was the
453 power consumption in the FO process for the dilution of brine reject using seawater as FS.
454 The prefiltration step is more demanding in terms of energy consumption than the FO process
455 itself. Generally, the overall energy demand in the FO process using TSE with filtration of the
456 FS and the DS is considered low and efficient in terms of cost-saving perspective. It is
457 noteworthy that the specific power consumption of the FO pretreatment is almost 142 times
458 lower than the nanofiltration pretreatment of seawater to MSF plant (Altaee et al., 2013)

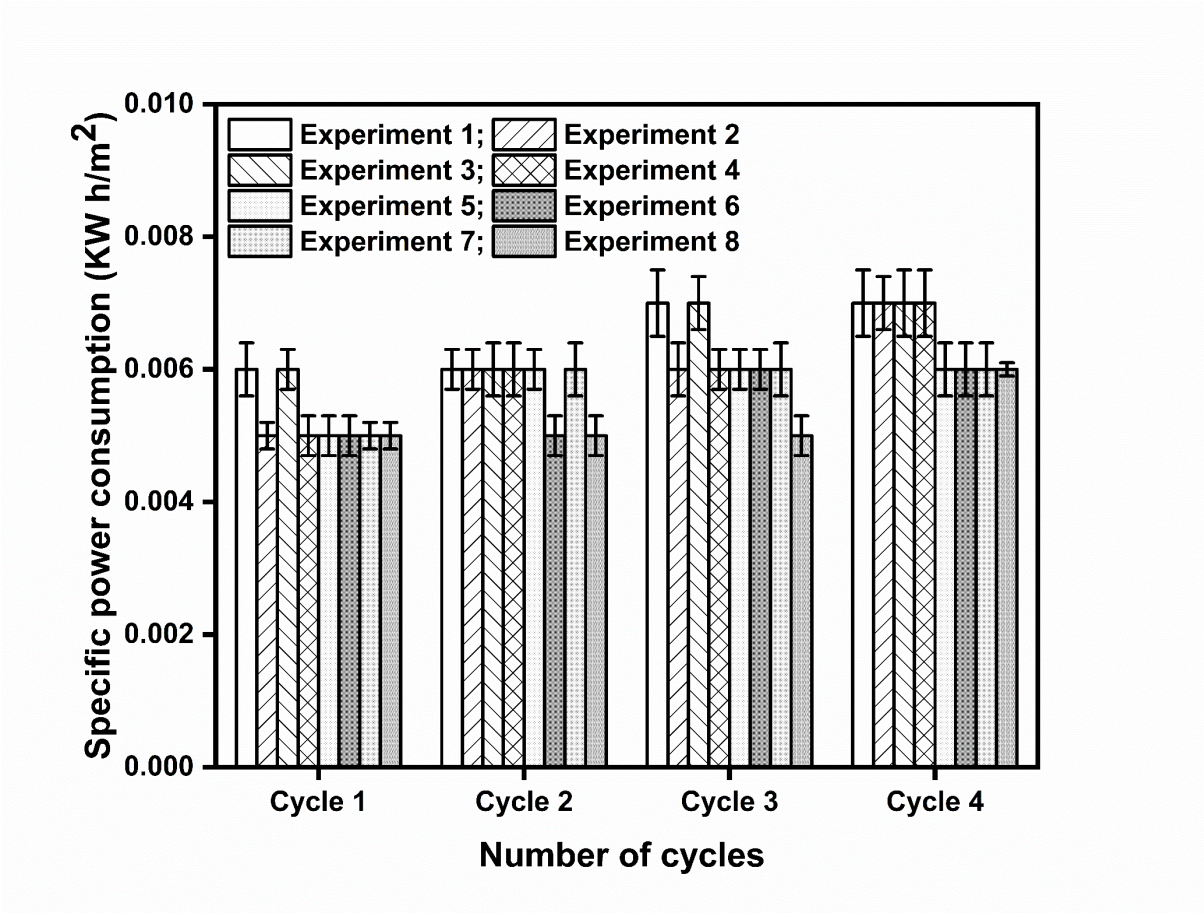

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Fig. 9. The performance of the FO experiments was conducted in terms of specific power consumption. (1-4) FO process without stream solution prefiltration, (5-8) FO process with prefiltration of FS & DS.

The membrane area required for brine reject pretreatment with seawater and TSE was calculated, assuming a capacity for a desalination plant of 10000 m3/day. The cost of Hydration Technology Innovation (HTI) FO element 8040FO-FS-P is USD 1719 per element, and it has a 16.5 m^2 active membrane area (Altaee et al., 2014b). The results were compared to the FO process using seawater as a feed solution (Khanafer et al., 2021). The FO membrane area was slightly increased, ~2%, when the membrane orientation changed from AL-FS to AL-D, whereas there was a 363% increase in the FO membrane area when the membrane orientation switched from AL-DS to AL-FS due to the sharp decrease in the water flux (**Table 3**). The drop in water flux is caused by the severe concentration polarization when seawater was the FS. Compared to the FO process used seawater feed solution, the results demonstrated that using TSE feed solution decreased the number of FO elements significantly. Using the TSE feed solution led to a 276% and 473% decrease in the number of FO elements required in the FO process, depending on the FO membrane orientation. The results indicate a significant advantage and cost-saving when TSE is used as a feed solution in the FO process.

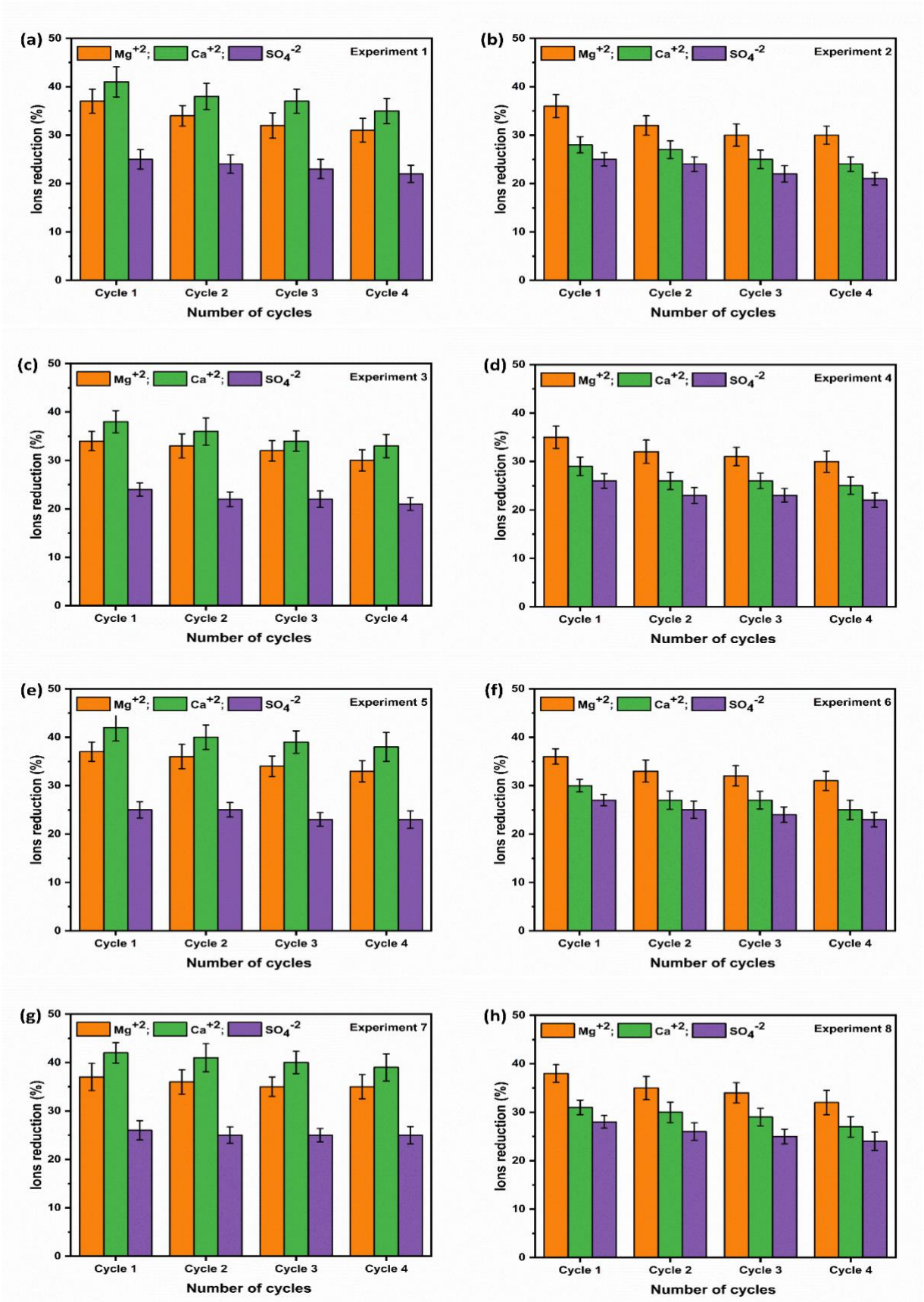
Table 3: Membrane area and cost for TSE and Seawater feed solutions in the FO process as pretreatment for the MSF plant.

Type Feed	Membrane orientation	Membrane area	No. FO Elements	Cost (USD)	Specific power (kWh/m^3)
TSE	AL-DS	12148	736	1265571	0.005
Seawater	AL-DS	33602	2036	3500733	0.02
TSE	AL-FS	11905	722	1240260	0.005
Seawater	AL-FS	56306	3413	5866093	0.03

3.7 Prefiltration and membrane cleaning effects on dilution of the Brine DS

The penetration of pure water throughout the FO membrane diluted the brine reject; therefore, a reduction in the concentrations of the ions occurs (Thabit et al., 2019). In this study, the concentrations of Mg^{2+}, Ca^{2+}, and SO_4^{2-} in the DS were studied in all FO treatment cycles, i.e. in both membrane orientations, with and without prefiltration. The concentration of Mg^{2+} and Ca^{2+} were measured using ion chromatography (7900 ICP-MS provided by Agilent technologies) and $SO4^{2-}$ using Dionex VWDIC manufactured by HPIC. **Fig. 10** showed the variation in the percentage of the concentration of the divalent ions from one cycle to another within the same experiment. Magnesium, calcium and sulfate ions are mainly responsible for scale formation in the MSF heat exchanger; for this reason, the reduction of these ions will help in scale control (Thabit et al., 2019). The ions reduction after each FO experiment is illustrated in **Fig. 10.** The percentage of ions reduction decreased due to the decline in water flux following the membrane cleaning within the same experiment. Overall, when the cleaning method's flow rate in the AL-DS orientation increased, it decreased the concentration of divalent ions of all three ions, where the reduction in Ca^{2+} is the highest. In the AL-FS tests, the reduction in Mg^{2+} and SO_4^{2-} was considerably greater than in the AL-DS, whereas the opposite for Ca^{2+}. The higher dilution of ions in the AL-FS test resulted from the greater average ware flux in these tests than AL-DS tests. For the same reason, in Exp1 (cycle 1), 37%, 41%, and 25 % were the percentage of reduction of Mg^{2+}, Ca^{2+} and SO_4^{2-}, respectively. These values decreased to 34%, 38%, and 24 % in Exp3 when the flow rate of the cleaning solution increased to 3 LPM**. (Fig. 10a & c)**. Cleaning the membrane with 3 LPM flow rate in the FO tests without the filtration of the solutions raised the initial water flux, which in turn caused irreversible fouling in the FO membrane and hence lower average water flux (**Fig. 3**);

thus, a dilution of the DS. The prefiltration of solutions in the FO tests provided better performance in the chain of the FO processes, decreasing membrane fouling in the subsequent filtration cycles 1 to 4. According to the results in Exp3 (3LPM cleaning without filtration) and Exp5 (with filtration), the reduction is higher after filtration; for example, in cycle 3 of Exp3, the reduction was 34 % compared to 39% in cycle 3 in Exp5 (AL-DS). The reduction was enhanced when an osmotic backwash was used; it counted 40% in cycle 3 (Exp7). In light of these results, the desired FO process would be in the AL-DS membrane orientation, filtration of feed and DSs, and cleaning with 40 °C DI water and osmotic backwash in order to tackle membrane fouling. Compared to the FO process performed with seawater FS, using wastewater FS achieved a 40% decrease in calcium concentration and 35% and 25% decrease in the concentrations of magnesium and sulfate, respectively (Exp7). This is more than 14%, the targeted dilution of the brine reject in an MSF plant operates at 112 °C (Morin, 1993). In effect, with the wastewater FS, the MSF plant could operate at top brine temperature higher than 112 °C and without antiscalant. Therefore, the FO process will improve the MSF plant's performance, avoid antiscalant use, and reduce the environmental pollution associated with the brine reject and wastewater disposal. Results revealed that the FO process has successfully diluted the brine using wastewater as FS.



521 **Fig. 10.** Reduction of Mg^{2+}, Ca^{2+} and SO_4^{2-} in % in FO experiments, (**a-d**) without prefiltration
522 and (**e-h**) with prefiltration.

4. Conclusions

In this study, treated wastewater and brine-reject were the FS and the DS, respectively, in a FO system. The performance of the FO process was correlated to the operating parameters studied. Experimental work showed that the prefiltation of the stream solutions was effective in terms of better water flux and fouling mitigation. The average water flux generated was considerably higher in the AL-FS mode in all cycles following the physical cleaning. The maximum average water flux achieved was 31.1 L/m^{2}h when the prefiltered wastewater was the FS compared to 9.57 L/m^{2}h when real seawater was used as FS. The physical cleaning methods reduced the membrane fouling and restored the water flux to a minimum of 86% at the end of cycle 4 of the FO experiment. SEM/EDS and FTIR analysis revealed that cleaning with 40 °C DI water and NaCl osmotic backwash effectively reduced the fouling that was believed to be reversible and not severe. The dilution of the brine reject solution was successfully achieved with the reduction of the ions reached 43%. The proposed FO system consumed maximum energy of 0.007 kW h/m^2, which is a promising economical outcome in competition with 0.020 kW h/m^2 consumed when seawater was used as FS. Overall, the high water flux recorded, the efficiency of the cleaning methods used, and the potential to reduce the divalent ions in the DS are the results that can build up further research. The outcomes of this study revealed the potential of the FO process in the dilution of brine-reject using wastewater and 40 °C DI water with NaCl osmotic backwash for membrane cleaning. There is about a 473% decrease in the number of FO elements and membrane cost when the TSE replaces seawater as the feed solution in the FO process. The FO process will reduce concentrated brine discharge to seawater, chemicals use and prevent thermal pollution due to brine discharge at 40 °C to seawater. The promising results of this study open more doors

for additional research on implementing the FO process in wastewater application for brine reject dilution for reuse in the MSF plant.

Appendix A

Table S1. Summary of the operational conditions applied in the experiments conducted in this study.

Experiment (Exp)	Membrane orientation	Cleaning (30min,40°C)		FS & DS treatment
		Active layer	Support layer	
1	AL-DS	DI water, 2LPM	DI water, 2LPM	-
2	AL-FS	DI water, 2LPM	DI water, 2LPM	-
3	AL-DS	DI water, 3LPM	DI water, 3LPM	-
4	AL-FS	DI water, 3LPM	DI water, 3LPM	-
5	AL-DS	DI water, 3LPM	DI water, 3LPM	20 µm filter
6	AL-FS	DI water, 3LPM	DI water, 3LPM	20 µm filter
7	AL-DS	DI water, 3LPM	45g/L NaCl, 3LPM	20 µm filter
8	AL-FS	45g/L NaCl, 3LPM	DI water, 3LPM	20 µm filter

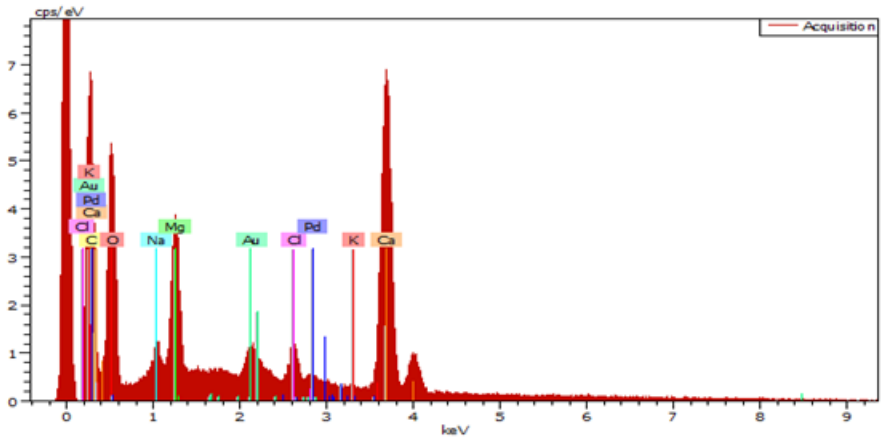


Fig. S1. Associated EDS spectrum of the sample membrane in the SEM-EDS analysis.

Table S2. Table showing the elements of the spectrum in numbers in the SEM-EDS analysis.

Spectrum: Acquisition

El	AN	Series	un. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
C	6	K-series	11.80	17.68	31.89	1.52
O	8	K-series	18.89	28.29	38.31	2.39
Na	11	K-series	0.50	0.74	0.70	0.06
Mg	12	K-series	3.28	4.91	4.38	0.20
Cl	17	K-series	1.25	1.87	1.14	0.07
K	19	K-series	0.04	0.06	0.03	0.03
Ca	20	K-series	28.50	42.69	23.08	0.98
Pd	46	L-series	0.41	0.62	0.13	0.05
Au	79	M-series	2.10	3.15	0.35	0.11
		Total:	66.77	100.00	100.00	

Fouling studies:

An HP4750 dead-end setup (Sterlitech USA) was used to study the antifouling properties of Porifera TFC membrane with wastewater as model foulant. Experiments were performed using both active and SL facing FS. First, pure water flux (J1) was measured for an hour at 3 bar pressure. Later wastewater was used as feed, and permeate flux (JP) was calculated for an hour. The used membrane was washed with DI water before measuring pure water flux (J2) again. Similar experiments were performed for four cycles. Reversible (Rr), irreversible (Rir), total fouling (Rt) and flux recovery ratio was calculated using equation 1, 2, 3 and 4, respectively.

$$R_r\ (\%) = \left(\frac{J_2 - J_P}{J_1}\right) \times 100 \quad (1)$$

$$R_{ir}\ (\%) = \left(\frac{J_1 - J_2}{J_1}\right) \times 100 \quad (2)$$

$$R_t\ (\%) = \left(1 - \frac{J_P}{J_1}\right) \times 100 \quad (3)$$

$$FRR\ (\%) = \left(\frac{J_2}{J_1}\right) \times 100 \quad (4)$$

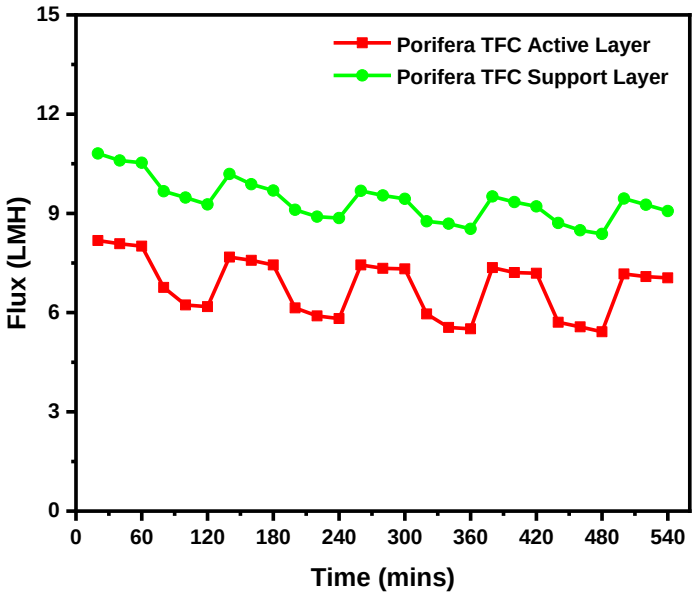


Fig. S2. Pure water flux and wastewater flux measurements for Porifera TFC active and SL membrane.

Table S3. Reversible, irreversible, total fouling and flux recovery ratio calculated for the Porifera TFC membrane.

	Reversible fouling (R_r) %	Irreversible fouling (R_{ir}) %	Total fouling (R_t) %	Flux Recovery Ratio (FRR) %
AL - Cycle 1	11.24	6.11	17.35	93.88
AL - Cycle 2	15.89	9.04	24.94	90.95
AL - Cycle 3	17.11	10.02	27.14	89.97
AL - Cycle 4	17.84	12.34	30.02	87.65
SL – Cycle 1	4.81	5.73	10.55	94.26
SL – Cycle 2	5.27	10.45	15.73	89.54
SL – Cycle 3	6.93	12.02	18.97	87.97
SL – Cycle 4	7.03	12.58	19.62	87.41

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