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Towards sustainable circular brine reclamation using seawater reverse osmosis, membrane distillation and forward osmosis hybrids: An experimental investigation

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Abstract

Desalination and wastewater treatment technologies require an effective solution for brine management to ensure environmental sustainability, which is closely linked with efficient process operations, reduction of chemical dosages, and valorization of brines. Within the scope of desalination brine reclamation, a circular system consisting of seawater reverse osmosis (SWRO), membrane distillation (MD), and forward osmosis (FO) three-process hybrid is investigated. The proposed design increases water recovery from SWRO brine (by MD) and dilutes concentrated brine to seawater level (by FO) for SWRO feed. It ultimately reduces SWRO process brine disposal and improves crystallization efficiency for a zero-liquid discharge application. The operating range of the hybrid system is indicated by a seawater volumetric concentration factor (VCF) ranging from 1.0 to 2.2, which covers practical and sustainable operation in full-scale applications. Within the proposed VCF range, different operating conditions of the MD and FO processes were evaluated in series with concentrated seawater as well as real SWRO brine from a full-scale desalination plant. Water quality and membrane surface were analyzed before and after experiments to assess the impact of the SWRO brine. Despite their low concentration (0.13 mg/L as phosphorous), antiscalants present in SWRO brine alleviated the flux decline in MD operations by 68.3% compared to operations using seawater concentrate, while no significant influence was observed on the FO process. A full spectrum of water quality analysis of real SWRO brine and Red Sea water is made available for future SWRO brine reclamation studies. The operating conditions and experimental results have shown the potential of the SWRO-MD-FO hybrid system for a circular brine reclamation.

Keywords: Seawater reverse osmosis; Membrane distillation; Forward osmosis; Brine reclamation; Hybrid desalination; Scaling inhibition

1 Introduction

Intensive human activities led to significant natural resources depletion and pollution spread across environmental ecosystems (WWAP and UNESCO, 2019). Water treatment and desalination technologies have experienced tremendous growth with imminent challenges to cover the exponential growth of water demand (Elsaid et al., 2020; Iqbal et al., 2021). Nevertheless, producing high-quality water still faces a number of constraints. One of the most common issues is the discharge of high concentration brine from water treatment plants (e.g., municipal, industrial, mining wastewater treatment plants, and seawater desalination plants) (Eke et al., 2020). These discharges adversely affect the marine environment and significantly slow the self-renewal of natural ecosystems (Liu et al., 2018; Petersen et al., 2018), and regulations on brine discharges has been updated to minimize the environmental impact of these harmful streams (Kress, 2019).

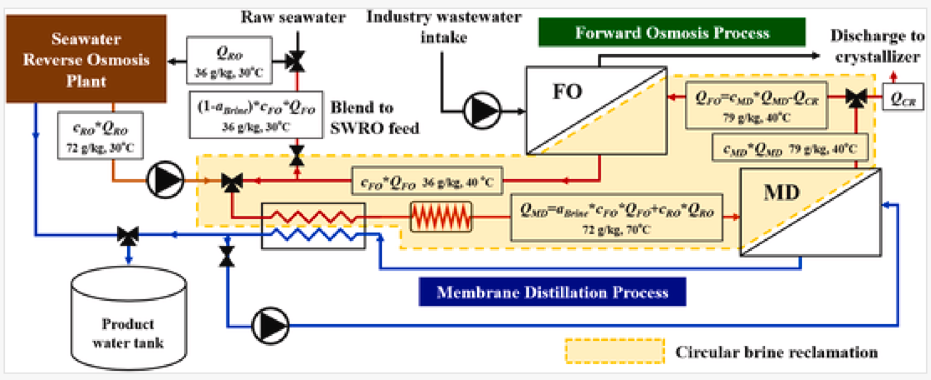
Minimization and recycling of concentrated brine have received great attention as key solutions for achieving sustainable desalination and have been sought through the integration of different technologies and the synergies of individual processes (Muhammad and Lee, 2019; Wagner et al., 2020). Despite all efforts on sustainable brine management, cases implemented in full-scale plants are rarely reported. One example is the Fujairah II desalination plant, which is the largest seawater reverse osmosis and multi-effect distillation (SWRO-MED) hybrid process, delivering 591,000 m^3/day of water (Lienhard et al., 2016). In this plant, the brine discharged from the SWRO process is recycled as a feed to MED. However, the impact on brine reclamation and higher water recovery is constrained by MED limitations, such as vulnerability to metal corrosion and scaling at high salinity feeds (Lienhard et al., 2016). Since SWRO process has become a major desalination technology in recent decades due to its high energy efficiency and high water recovery compared to other conventional processes; SWRO hybrid systems can pave the way for economic viability and environmental sustainability in minimizing the disposal of brine (Park et al., 2019; Sommariva, 2017).

To overcome the challenges of SWRO, emerging technologies (e.g., membrane distillation (MD), forward osmosis (FO), and nanofiltration (NF)) are being adopted to build SWRO hybrid systems, such as NF-RO-MED, NF-RO-ED, and FO-RO-ED (Ahmed et al., 2020). MD is a membrane process based on thermal-driven separation. It allows freshwater to be produced from concentrated brine through the hydrophobic membrane, resulting in a high salt rejection rate (i.e., 99.9% for non-volatile solute) (Deshmukh and Elimelech, 2017; Soukane et al., 2019). The process is less sensitive to feed salinity compared to other membrane-based desalination processes since water crosses the membrane in a vapor state due to the difference in partial vapor pressures between feed and permeate sides of the membrane. Therefore, the MD process represents a potentially efficient solution to treat RO brine. On the other hand, in an FO process, the osmotic pressure gradient generated by the concentration difference between draw and feed solutions (DS and FS) is utilized as a driving force by using a hydrophilic membrane (Vu et al., 2019). Hence, water molecules transfer from the feed solution (i.e., higher water activity) to the draw solution (i.e., lower water activity) leading to high rejection of solutes (Kim et al., 2021). In this process, dilution and concentration occur simultaneously in the draw and feed sides, respectively, and the diluted draw solution can be recycled as feed for desalination processes such as RO and MD (Phillip et al., 2010). Moreover, potential of MD and FO integration has been reported in previous studies (Ang et al., 2020; Son et al., 2021) to maximize a system performance. In MD-FO hybrid system, MD regenerates the DS of FO process and FO produced water from impaired wastewater with low salinity (Kim et al., 2019; Li et al., 2020).

Therefore, in this study, a sustainable design of SWRO-MD-FO hybrid system (see Fig. 1) is proposed to (i) minimize brine disposal, (ii) maximize water recovery with MD, and (iii) reuse SWRO brine with FO as DS in a circular brine reclamation system. As with the SWRO-MED hybrid mentioned previously (Lienhard et al., 2016), integrating MD with SWRO has a high potential for sustainable operation in a full-scale desalination plant. In addition, the consecutive FO process is also fully integrated for brine reclamation. Different authors considered the separate use of MD and FO processes to handle SWRO brine (Duong et al., 2015; Ge et al., 2014; Gryta, 2012), but to the best of our knowledge, integration of SWRO-MD-FO is not yet investigated, probably due to a complex process combination. The main challenge in such a system is membrane scaling and fouling at the recycling DS side. The inorganic crystal formation at the operating conditions (i.e., temperature and concentration) above limits will affect system sustainability. For a practical understanding of the hybrid system operation, individual processes are studied in-series with real water samples since chemicals in real SWRO brine play significant roles in both MD and FO processes (Gwak and Hong, 2017; Zhang et al., 2015). Therefore, in this study, the sustainable operating range of the SWRO-MD-FO hybrid system is proposed, and fouling tendency of the MD and FO processes with antiscalant presence are investigated by using real brine samples collected from a full-scale commercial SWRO plant.

alt-text: Fig. 1

Fig. 1



Process flow diagram of the SWRO-MD-FO hybrid system for brine reclamation. c_{RO}, c_{MD}, and c_{FO} represent concentration ratios for SWRO, MD, and FO, respectively. a_{Brine} is the fraction of diluted brine from FO redirected to SWRO.

2 SWRO-MD-FO hybrid system for circular brine reclamation

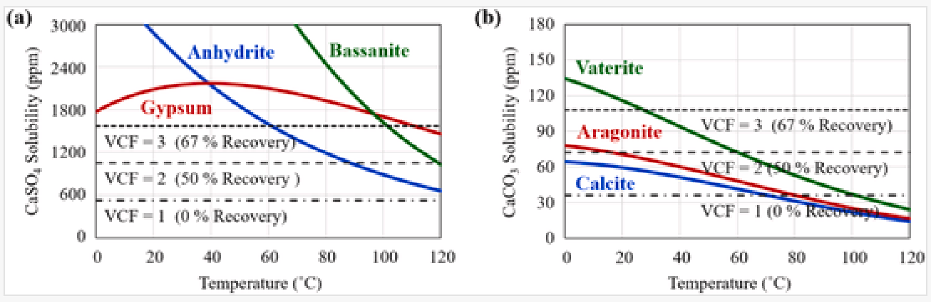
The membrane-based hybrid desalination system shown in Fig. 1 is proposed to minimize brine disposal and achieve the flexible operation of an integrated system. Three membrane based processes (SWRO, MD, and FO) can be operated with different parameters (i.e., recovery, and blending ratio). However, search for optimum parameters is not covered in this study and focus is set on the fouling phenomena under the proposed operating conditions. In the circular brine stream, the SWRO brine goes through consecutive concentrating and diluting processes. The SWRO brine is heated before entering the MD process by low-grade thermal energy (i.e., waste heat) (Ghaffour et al., 2019). The concentrated MD effluent (MD brine outlet) enters the FO process as DS with a temperature of 40 °C, typical of FO process inlet conditions (Adhikary et al., 2020; Xie et al., 2013). The highly concentrated DS is diluted through FO permeation from impaired wastewater, until a typical seawater concentration (~35 g/L salinity) is reached. This effluent can be blended with SWRO feed in a continuous mode, and also can be recirculated within the loop depending on the operating conditions.

The concept of SWRO-MD-FO hybrid system is straightforward. However, for practical applications it requires to find the sustainable operation parameters and to understand the impact of chemicals present in the brine on the performance of MD and FO processes. It is necessary to comprehend the limitations of standalone MD and FO processes according to salt solubility of all individual streams. Therefore, an optimized operating mode of the SWRO-MD-FO hybrid system should consider (i) the solubility of existing inorganics and (ii) the feasible range of MD water recovery. To capture the dynamic changes of brine salinity in the hybrid system, the seawater volumetric concentration factor (VCF), a normalized value of seawater concentration, is used as a primary variable to define operating conditions. The VCF is based on the seawater concentration of 36 g/kg considering available real SWRO brine sample used in the experiments and previous literature (Eke et al., 2020; Elimelech and Phillip, 2011).

In seawater desalination systems, calcium sulfate and calcium carbonate are the major inorganic fouling contributors. (Zhang et al., 2015). The solubility curves of different structural forms of calcium sulfate and calcium carbonate are presented as a function of temperature in Fig. 2 using data from the literature (Al-Rawajfeh et al., 2012; Al Rawahi et al., 2017; Choi et al., 2019; Raharjo et al., 2016; Van Driessche et al., 2017). Depending on the degree of hydration, calcium sulfate exists in three main forms, i.e., gypsum (the dihydrate, $CaSO_4 \cdot 2H_2O$), bassanite (the hemihydrate, $CaSO_4 \cdot 0.5H_2O$) and anhydrite ($CaSO_4$) (Van Driessche et al., 2017; Zarga et al., 2013). Therefore, to reduce the risk of severe scaling, membrane processes are operated below the solubility curves or by employing antiscalants. Since the solubility limit of the anhydrite at 80 °C corresponds to a VCF of 2.3, the proposed range of sustainable operation does not exceed a VCF of 2.2.

alt-text: Fig. 2

Fig. 2



Solubility curves of (a) calcium sulfate, and (b) calcium carbonate with respect to VCF and seawater temperature.

Whereas calcium carbonate can be found in the environment in its various forms, including anhydrous (e.g., calcite, aragonite, and vaterite) and hydrated (e.g., monohydrocalcite and ikaite). In seawater, aragonite and calcite are dominant polymorphs of calcium carbonate (Elfil and Roques, 2001; Marion et al., 2009). However, in desalination systems, calcium carbonate scaling is considered as soft scaling, which is more reversible compared to sulfate scaling (Rahmawati et al., 2012; Warsinger et al., 2015). Hence, calcium sulfate scaling is mainly considered in defining the operation range.

In the hybrid system, MD limits the process as it regulates the operation range by controlling the FO DS concentration. Theoretical limitations of MD recovery remain below 10% in the practical operating range of GOR (40–100%), as illustrated in Fig. S1 for different ranges of temperature differences between feed and permeate. The experimental results of pilot operations taken from the literature show that the practical MD recovery is constrained to less than 8% (Andrés-Mañas et al., 2020; Dutta et al., 2020; Lee et al., 2020; Najib et al., 2019; Ruiz-Aguirre et al., 2017, 2018, 2018; Schwantes et al., 2013, 2018; Winter et al., 2011, 2012), and the potential range of the hybrid system in this investigation is selected accordingly. As such, the VCF does not exceed 2.2, which represents 10% concentration increase by MD from a VCF of 2.0. The latter corresponding to a maximum 50% recovery of the SWRO process (VCF increase from 1 to 2). The 10% recovery of MD can be considered as additional water production using available thermal energy sources (i.e., low-grade waste heat) on-site to maximize production. The range of targeted VCF is reported in Table 1, and the detailed compositions of seawater and SWRO brine used in this study are reported in Table 2.

alt-text: Table 1

Table 1

Operation range of VCF in the SWRO-MD-FO hybrid system with SWRO brine.

Process Description	Range of VCF
Raw Seawater	1.0
Seawater Reverse Osmosis	from 1.0 to 2.0
Membrane Distillation	from 2.0 to 2.2
Forward Osmosis	from 2.2 to 1.0

alt-text: Table 2

Table 2

Water quality of SWRO brine and seawater concentrate before/after MD operation at different temperatures.

* Ion concentrations in mg/L	Silicon	Phosphorous	Fluoride	Boron	Bicarbonate	Sulfate	Chloride	Calcium	Potassium	Magnesium	Sodium	TDS (mg/L)	Cond (μS/cm)	TOC(mg/L)	pH		
	0.11	0.13	0.71	4.49	277.3	5326.2	40,980	713.3	1034.1	2128.5	21,432	71,827	90,625	2.36	8.17	Initial	Brine, SWRO
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.02	0.00	0.02	0.16	0.8	59.3	443	7.4	23.8	23.0	138	282	646	0.39	0.10		
	0.23	0.15	0.75	5.17	296.8	5812.0	44,359	761.7	1086.5	2216.9	22,966	77,505	97,450	2.54	8.15	MD @80°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.03	0.02	0.01	0.23	1.6	2.5	225	0.7	53.0	132.1	159	466	50	0.11	0.10		
	0.28	0.14	0.69	5.18	283.4	5659.4	44,040	769.0	1089.9	2234.1	23,546	77,627	97,200	2.47	8.27	MD @70°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.03	0.03	0.05	0.10	3.9	182.0	802	15.0	85.7	187.9	77	809	100	0.05	0.07		
	0.23	0.15	0.73	5.24	289.2	5918.3	45,136	771.8	1145.7	2292.7	23,642	79,202	99,300	2.77	8.02	MD @60°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.16	0.01	0.04	0.23	1.1	24.9	168	15.2	36.2	146.3	49	366	200	0.15	0.04		
	0.32	0.14	0.71	5.18	291.7	5889.0	45,044	786.3	1135.1	2318.5	23,960	79,430	98,950	2.59	8.10	MD @50°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.03	0.01	0.06	0.08	0.7	1.4	155	11.7	66.9	157.5	18	241	150	0.08	0.02		
	0.21	0.03	0.73	8.05	235.9	5635.7	40,601	746.0	1120.7	2321.0	21,278	71,888	90,825	3.03	8.23	Initial	Concentrate, Seawater
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.04	0.02	0.02	0.24	1.3	34.6	419	10.2	34.5	51.6	98	284	444	0.86	0.09		
	0.56	0.03	0.69	9.46	258.3	6374.9	46,033	811.6	1254.4	2507.6	23,663	80,913	100,750	3.31	8.16	MD @80°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.08	0.02	0.07	0.22	2.1	8.3	221	8.4	30.3	144.4	149	189	950	0.01	0.06		
	0.44	0.04	0.76	9.54	276.8	6503.8	46,917	816.9	1349.9	2605.0	24,447	82,927	102,100	3.27	8.29	MD @70°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.04	0.03	0.01	0.41	1.7	26.2	238	26.1	23.9	118.0	84	349	700	0.08	0.02		
	0.50	0.02	0.75	9.95	253.0	6052.7	43,907	798.5	1248.0	2497.6	23,499	78,267	98,950	3.20	8.09	MD @60°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.04	0.01	0.06	0.31	2.3	170.6	958	9.5	29.3	143.4	200	862	150	0.09	0.02		
	0.50	0.03	0.66	9.78	267.0	6457.4	46,663	808.1	1346.8	2523.1	24,144	82,220	101,800	3.37	8.03	MD @50°C	
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±		
	0.08	0.02	0.03	0.14	1.1	28.4	326	9.9	52.4	174.9	234	198	700	0.05	0.07		

The actual brine was obtained from full-scale SWRO desalination plant at King Abdullah University of Science and Technology (KAUST), Saudi Arabia (Belila et al., 2016), and concentrated Red Sea water was employed as a reference to be compared with the brine. The lab-scale MD and FO experiments were conducted at different MD feed temperatures and the fouling (or anti-fouling) potential of the existing chemicals in the brine was assessed at the proposed operating conditions as described in the sequel.

3 Materials and methods

3.1 KAUST SWRO desalination plant

A full-scale SWRO desalination plant, supplied from the Red Sea, provided the brine used in this study. It is located at KAUST, Saudi Arabia, and has a capacity of 40,000 m^3/day. The plant uses conventional pre-treatment processes with spruce media filter (SMF), a two-pass RO system (SWRO and BWRO) (Rahmawati et al., 2012) to control fouling and scaling, and a post-treatment system (see Fig. S2). Antiscalant is added (1.5 mg/L of phosphate based product) to mitigate membrane fouling of RO process.

The (1st pass) SWRO process is operated to maximize water recovery, reaching salt rejection of 99.75%. Part of SWRO product water is supplied to the brackish water RO process (BWRO, 2nd pass) after injecting caustic soda and antiscalant. In this study, the real brine sample for the experiments is collected from the reject of the 1st pass SWRO process. Additional details on the operating mode are presented in the Supplementary Information document (Section S2).

3.2 Membrane distillation (MD) and forward osmosis (FO) experiments

Commercially available polytetrafluoroethylene (PTFE) and polyamide (PA) thin-film composite (TFC) membranes were utilized in the MD and FO processes, respectively (Ghaffour et al., 2019; Lee et al., 2018; Lee and Ghaffour, 2019), and automated low-pressure membrane experimental set-up is customized for both MD and FO experiments. Detailed descriptions are presented in the Supplementary Information document (Section S3).

In order to investigate the feasibility of the proposed hybrid system, MD and FO experiments were conducted in series and in standalone mode, after which water quality and membrane surfaces were analyzed. Real brine, collected from KAUST SWRO desalination plant or from concentrated seawater was used as MD feed, and the concentrated MD feed was utilized as a draw solution for the FO process to demonstrate in-series operation. Samples from a full-scale plant are more representative of the complex nature of real SWRO brines compared to brine samples prepared by adding antiscalant to seawater. The seawater concentrate was prepared as a reference by evaporating Red Sea water at 25 °C using a rotary evaporator (Laborota 20 control, Heidolph Instruments) equipped with a vacuum pump (PC 3004 VARIO, Vacuubrand).

As discussed in Section 2, the target water recovery of MD was set at 10% (up to a VCF of 2.2). MD experiments were performed in DCMD configuration with inlet temperatures of 50, 60, 70, and 80 °C. The upper limit of the feed temperature (80 °C) is set following the solubility limit of calcium sulfate (Fig. 2) and the integration with low-grade thermal energy (70–90 °C) (Ghenai et al., 2021; Tong et al., 2020). The inlet permeate temperature was set to 30 °C, and the flow rate was set to 0.5 L/min for both MD feed and permeate streams. The concentrated MD feed effluents were cooled before entering the FO process as DS. The target temperature of concentrated feed from MD is set to 40 °C, which is a feasible value given an MD permeate at 30 °C (Khalifa et al., 2017), while the FO feed temperature was set to 30 °C. During FO experiments, DS circulates in batch mode until its concentration reaches the VCF of 1.0, which indicates a change of VCF from 2.2 to 1.0. The active layer of FO faced the feed solution (AL-FS) for the FO process. A flow rate of 0.5 L/min for both feed and draw was applied in all experiments. Analytical methods of experiments are presented in the Supplementary Information document (Section S4).

4 Results and discussion

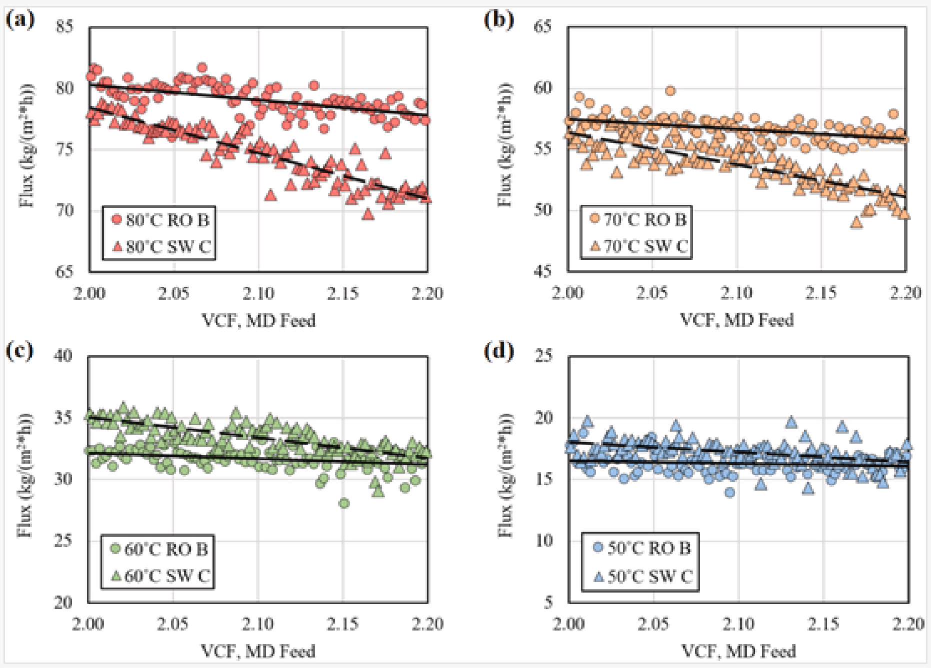
4.1 Evaluation of the membrane distillation (MD) process

The MD process performance was first investigated to assess the feasibility of the proposed SWRO-MD-FO integration. For membrane processes, fouling is the main challenge to overcome, and it hinders the sustainable operation of the MD process as well. Inorganic scaling in MD is the main concern when feed is saline brine (Mericq et al., 2010; Tijing et al., 2015; Yan et al., 2017). Therefore, the influence of residual chemicals (i.e., antiscalant) present in the real SWRO brine on the performance of the membrane processes (MD and FO) stands among the main interests of this investigation. For MD operations, two separate sets of experiments with (i) real SWRO brine (RO B) and (ii) Red Sea water concentrate (SW C) were conducted at different temperatures with the same initial VCF of 2.0.

The influence of the antiscalant present in SWRO brine can be seen from the variation of MD flux with respect to VCF (Fig. 3). As the VCF increased from 2.0 to 2.2, the initial flux decreased by 9.5% with seawater concentrate, while only a 3.2% reduction with SWRO brine was observed under the same experimental conditions. When operating with SWRO brine, the absolute value of flux reduction with increasing VCF was accelerated at higher temperature, but the decrease in permeation flux was similar, with an average value of 9.2% and a standard deviation of 0.29%. Moreover, the difference in flux decline between SWRO brine and SW concentrate has been shown to increase with higher MD feed temperature operation. This observation indicates that calcium scaling is involved in the reduction of permeation flux, as the solubility limits of calcium carbonate and calcium sulfate decrease at higher temperatures (Fig. 2). The qualitative assessment of foulants is discussed using the SEM and EDX analysis in Section 4.4.

alt-text: Fig. 3

Fig. 3



MD fluxes versus VCF using SWRO brine (RO B) and seawater concentrate (SW C) at different temperatures, (a) 50 °C, (b) 60 °C, (c) 70 °C, and (d) 80 °C.

Inorganic scales in MD and other membrane processes usually occur via two pathways; (i) bulk, or (ii) surface crystallization. Bulk crystallization develops from nuclei into a bulk crystal in the supersaturated solution. On the other hand, a deposited nuclei on the membrane surface initiate surface crystallization, which causes pore-clogging rather than formation of a surface layer (Choi et al., 2019; Tijing et al., 2015). This typical flux decline observed in our experiments can be attributed to particle-induced fouling from the bulk solution rather than foulant growth on membrane surfaces (Gilron et al., 2013). It implies that fouling which occurs with seawater concentrate is mainly a result of bulk crystallization, enhanced by rapid changes in solubility.

On the other hand, the compilation of all experiments gives an average difference of 5.4% between initial MD fluxes using seawater concentrate and those with SWRO brine due to their similar ionic composition. At a glance, at a feed temperature of 80 °C, initial fluxes using SWRO brine and using seawater concentrate were found to be 80.3 kg/(m^2·h) and 78.4 kg/(m^2·h), respectively, which represents a 2.4% difference. VCF variations over time are shown in Fig. S4 to present the overall operation time and production rate as an additional information.

The positive impact of the antiscalant remaining in the brine has been observed in this study, although the behavior of antiscalant can differ widely depending on the quantity and type used (Duong et al., 2015; Ge et al., 2014; Gryta, 2012; Tijing et al., 2015). According to Peng et al. (2015), four different antiscalants could mitigate flux decline in MD operation at feed temperature of 77 °C, without membrane wetting. Another study showed that the presence of antiscalant in real MED brine had a beneficial effect on MD flux stability by prolonging the scale formation time, however, higher antiscalant concentrations led to a decrease of the initial flux (Elcik et al., 2020). On the other hand, the fouling potential of antiscalant in MD also has been reported by Tijing et al. (2015). It is explained that the degraded antiscalant cannot alleviate fouling, and can be diverted into the foulant during high-temperature MD operation. Thus, there is particular emphasis in the literature on an optimal antiscalant concentration in an MD feed. The feasibility of the hybrid system can be addressed when the concentration of antiscalant in real SWRO brine is favorable to sustainably operate the MD process. In the present study, the antiscalant concentration of the brine was lower than the concentration triggering membrane fouling and the antiscalant behaves as a scaling inhibitor in the MD process.

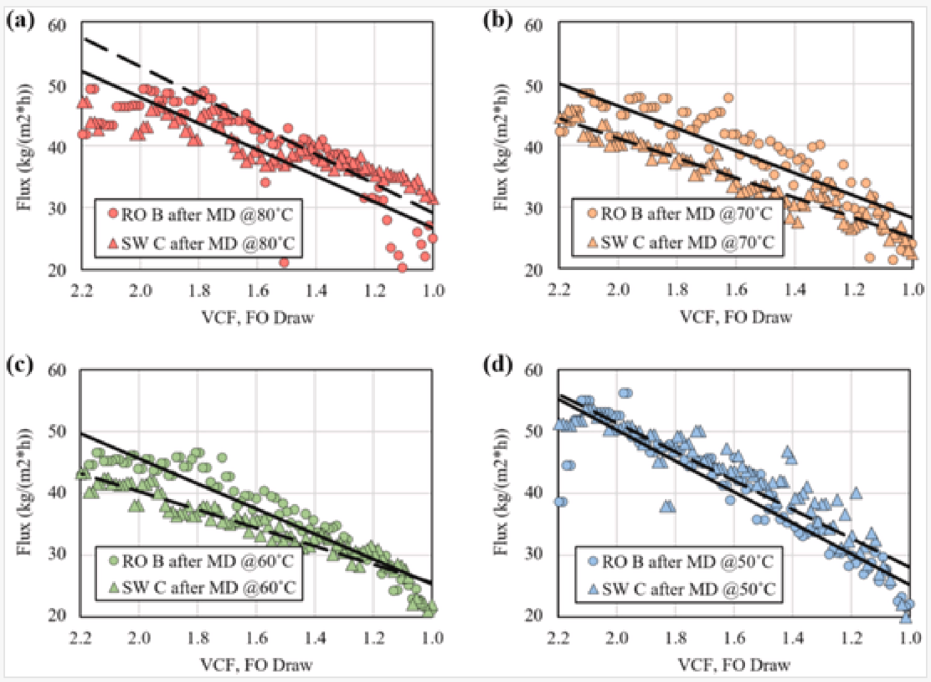
4.2 Evaluation of the forward osmosis (FO) process

FO experiments are exploited to evaluate the antiscalant influence with the concentrated (at different temperatures) MD feed as DS. The range of VCF was set to 2.2–1.0, and experiments using SWRO brine were compared to those with seawater concentrate. In the proposed hybrid, low-saline wastewater can be used as the FO feed. However, freshwater (10–15 µS/cm conductivity) was used instead to assess the actual impact of existing antiscalants on real SWRO brine, regardless of FO feed water quality.

FO flux results for VCFs in the range of 2.2–1.0 indicate that no antiscalant effect was observed when using SWRO brine in the FO operations. Neither positive nor negative meaningful impacts were observed, as shown in all FO fluxes trends (Fig. 4). The average values of the initial FO fluxes with the SWRO brine and seawater concentrate were close (51.8 and 50.3 kg/m^2/h respectively), and the standard deviation was less than 10% for all experiments, with both DS. The decline rates also lie within a similar range where the initial FO fluxes decrease by 41.0–54.5% in all experiments from 2.2 to 1.0 of DS VCF (55% dilution), with a standard deviation of 8.7%. The specific reverse solute flux (SRSF) values are measured as 0.136 g/kg and 0.133 g/kg for the SWRO brine and seawater concentrate, respectively. VCF changes during dilution are represented in Fig. S5, which was measured similarly in all FO experiments. As the osmotic pressure gradient was reduced, the dilution rate was mitigated over time.

alt-text: Fig. 4

Fig. 4



FO fluxes versus VCF for SWRO brine (RO B) and seawater concentrate (SW C) after MD operation. FO DS inlet temperature was set to 40 °C while MD was operated at different feed temperatures, (a) 50 °C, (b) 60 °C, (c) 70 °C, and (d) 80 °C.

For the FO process, the advantage of the SWRO brine was not evident in this study. However, its benefits are widely described elsewhere (Gwak and Hong, 2017; Lee et al., 2019) for it delays the scaling induction time and enhances osmotic driving force for FO. It can improve the FO operation of the proposed hybrid system if a challenging feed quality, such as high concentration wastewater, is used. Gwak and Hong (2017) presented the effect of antiscalant blended DS on the water flux and reverse salt flux (RSF). The study showed that antiscalant could inhibit scale formation via reverse diffusion of antiscalant. To maximize scaling mitigation in the proposed hybrid system, antiscalant can be added before the FO process and not just prior to SWRO. The addition of antiscalant is inevitable to maintain efficiency due to its degradation and loss. Moreover, for a practical implementation, the antiscalant characteristics have to be investigated as the influence of high-temperature required for MD operation on antiscalants vary depending on their type and properties (Peng et al., 2015). As reported by Li et al. (2015), six types of antiscalants have behaved distinctively in different dosages at high temperature (80 °C).

4.3 Effect of MD operating conditions on water quality

Water quality of the key streams was analyzed for an in-depth understanding of the rejection level of each process. Examining the antiscalant and its by-products concentration in real SWRO brine would be the ideal scenario to study antiscalant efficiency. However, the quantification of low-level chemicals in the real process or environment is still limited. Ion chromatography and UV spectrometry analyses are commonly used to quantify antiscalants (e.g., phosphates and phosphonates). However, measurements can be constrained by co-existing salts and organic matters, which makes the use of ICP method a more viable option (Armbruster et al., 2019; Baluyot and G. Hartford, 1996; Wang et al., 2019). In this study, the antiscalant present in real SWRO brine is investigated by the ICP-OES despite the limitation that it only measures overall value of phosphorous from all sources. However, the actual phosphorous from the antiscalant is estimated by subtracting the concentration of raw Red Sea water concentrate. Only water quality of MD feed and permeate was measured, of which results are reported in Table 2. The investigation of the FO process water quality is left for future work.

The seawater concentrate before experiments showed a TDS concentration (and conductivity) very close to those of the real SWRO brine (71,827 ± 282 mg/L and 71,888 ± 284 mg/L for the SWRO brine and seawater concentrate, respectively). The initial values in the table represent the quality of raw SWRO brine and seawater concentrate. The major differences concerned the concentrations of (i) phosphorous, (ii) boron, (iii) silicon, and (iv) total organic data. Among all, phosphorous concentration was significantly different, and was 4.3 times more abundant in SWRO brine than seawater concentrate. It confirms the presence of antiscalant (phosphate-based, phosphonate-based, or blended) in the real SWRO brine, but the amount is far less compared to concentrations reported in antiscalant studies (Gryta, 2012; Lee et al., 2019). Another interesting difference was the boron amount in the SWRO brine. Its concentration was 4.49 mg/L, which is only 57% of the amount found in seawater concentrate. A possible explanation is boron transport across SWRO membranes (Alpatova et al., 2018; Rahmawati et al., 2012). The proposed system recycles the diluted brine from the FO process with a minimum make-up seawater supply, which offers a more practical way to control boron in the product. The lower concentrations of silicon (0.11 mg/L) and TOC (2.36 mg/L) in real SWRO brine compared to those in seawater concentrate (0.21 mg/L and 3.03 mg/L) could be due to the pre-treatment process of the SWRO feed (Belila et al., 2016).

MD operation at temperatures between 50 and 80 °C did not bring a clear influence on the quality of the SWRO brine leaving the MD feed compartment. For most of ions, the percentage changes were measured to be within 1.5% differences. However, after MD operation, a perceptible change is noted in calcium proportion. The calcium percentage was reduced by 0.9% in SWRO brine experiments, but 3.8% of calcium percentage was deducted from the original percentage in seawater concentrate (before MD experiments). The result can be explained by the calcium related fouling in seawater concentrate experiments as it deposited on the membrane surface, and the bulk concentration of calcium became less than the SWRO brine in the MD experiments. The salt rejection of the MD process was measured as higher than 99.9% in all cases, and MD permeate conductivities were in the range of 10–15 μS/cm.

4.4 Evaluation of membrane fouling with respect to VCF changes

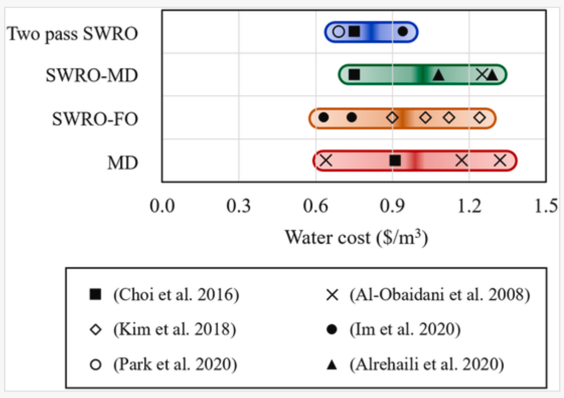
The membrane surfaces were analyzed to characterize the morphology and composition of fouling. MD membrane samples after SWRO brine and seawater concentrate experiments, at different temperatures (50–80 °C), were collected and prepared for SEM analysis. Data is presented in Fig. S6 with two different magnifications (x 1k and x 5k). No significant membrane fouling was noticed for both streams at a feed temperature of 50 °C, which is expected since experiments gave similar flux trend. However, at 80 °C, more fouling was observed with seawater concentrate (Fig. S6 (d)) than SWRO brine (Fig. S6 (b)). Not only quantities are different but morphology also exhibits distinct variation. The shape of foulants in the SWRO brine experiment are found as irregular aggregates, which is in contrast with the crystalline structure in seawater concentrate. For these two MD membranes, flux decline varied because of the difference in scale characteristics. Moreover, with seawater concentrate, fouling may have invaded pores with small particulates in the early stage of crystallization well before crystal development (Choi et al., 2019; Tijing et al., 2015).

Further analysis was conducted by EDX to identify the composition of foulants (Fig. S7). It shows that, using seawater concentrate, the crystal on the membrane surface was mainly calcium carbonate. The MD process is operated above the solubility limit of calcium carbonate **(**Fig. 2**)**, and its morphology is also consistent with previous studies (Alpatova et al., 2018; Guo et al., 2011; Huang et al., 2016). The two other foulants appearing with SWRO brine (Fig. S7 (d) and Fig. S7 (e)) are sodium chloride and complex aggregates, respectively. The antiscalant in SWRO brine influenced the foulant characteristics, although some inorganic foulants were observed on the MD membrane with SWRO brine operation. MD operation above 50 °C could lessen the efficiency of antiscalant, because antiscalants used in SWRO processes do not have a high thermal stability (Alpatova et al., 2018; Peng et al., 2015). In addition, the percentage of phosphorous found in the foulant using SWRO brine was significantly higher. Nevertheless, it is worth emphasizing that the fouling tendency presented is derived from changes of seawater VCF and not from long-term system operations, which needs to be considered for future research**.**

Since the SWRO-MD-FO concept is at the evaluation stage, a full cost analysis is not available yet. However, research related to other hybrids or standalone processes has been widely reported in various publications, of which results relevant to this investigation are summarized in Fig. 5 (Al-Obaidani et al., 2008; Alrehaili et al., 2020; Choi et al., 2016; Im et al., 2020; Kim et al., 2018; Park et al., 2020). From cost analysis of two pass SWRO, RO-MD, RO-FO, and standalone MD systems, the economic feasibility of the SWRO-MD-FO hybrid system is expected to fall within the lowest range, since additional recycling and treatment is provided.

alt-text: Fig. 5

Fig. 5



Water cost analysis of relevant technologies to SWRO-MD-FO hybrid.

5 Conclusions

The sustainable application of the proposed hybrid system was demonstrated with a circular brine reclamation through the SWRO, MD, and FO processes. Individual processes in the system can be designed for flexible operations according to target conditions. SWRO brine is further concentrated with the MD process and can be employed in-series as a DS for the FO process. The diluted brine (DS) by the FO process can be recycled to the main SWRO process (salinity similar to raw seawater) as a high quality feed. In addition, the system produces additional freshwater from the wastewater (FO feed) while the brine (DS) is diluted with the FO process. In this study, the MD process performance in the hybrid system is enhanced. Namely, the flux reduction rate over a VCF range from 2.0 to 2.2, was significantly mitigated with the RO brine as feed. FO experiments, carried out under different MD operating conditions, presented a stable operation at the same VCF range. Water quality analysis also confirmed the existence of antiscalants in the real SWRO brine, which inhibited salt crystallization. Calcium carbonate formation was found on the MD membrane surface when using seawater concentrate which was confirmed by the SEM and EDX analysis. The presence of antiscalant in SWRO brine mitigated invasive calcium related fouling, which generally causes a flux decline over VCF in MD operations. The experimental investigation with the real SWRO brine allowed to present the potential of the proposed hybrid system. Optimization of possible operating modes will enhance the sustainability of the SWRO-MD-FO hybrid system, which will be the focus of future research.

Declaration of competing interest

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

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Appendix A Supplementary data

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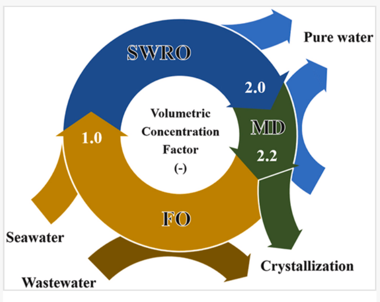
Graphical abstract

alt-text: Image 1



Highlights

- A flexible design of SWRO-MD-FO hybrid system is developed for brine recycling.
- Real SWRO brine from a full-scale desalination plant is treated.
- Feasibility of the proposed system is experimentally evaluated with MD and FO operations.
- Operation range of seawater VCF from 1.0 to 2.2 is experimentally investigated.
- Flux decline in MD is mitigated through retardation of $CaCO_3$ crystallization.

Appendix A Supplementary data

The following is the Supplementary data to this article:

Multimedia Component 1

Multimedia component 1

alt-text: Multimedia component 1

Queries and Answers

Q1

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