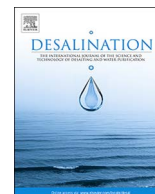




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Reverse osmosis brine treatment using direct contact membrane distillation: Effects of feed temperature and velocity

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ABSTRACT

Membrane distillation (MD) is a promising technology for reverse osmosis (RO) brine treatment due to its superb tolerance to high salinity. In this work, a hydrophobic PVDF membrane was applied to treat simulated RO brine under different feed temperatures and velocities with membrane flux, permeate conductivity and thermal performance monitored. The raw membrane was characterized with respect to contact angle, porosity, thickness, maximum pore sizes, liquid entry pressure and clean water flux. The fouled membrane was characterized by scanning electron microscope (SEM) coupled with an energy dispersive X-ray spectroscopy (EDX). The results showed that MD achieved excellent desalination performance with the permeate conductivity lower than $11 \mu\text{S cm}^{-1}$ and recovery rate higher than 70% during the RO brine treatment. Almost no membrane scaling was observed in the initial stage, but significant scaling occurred due to overconcentration (concentration factor > 3.3) of RO brine, resulting in serious pore wetting and decreased thermal performance. Moreover, increasing feed velocity was helpful to alleviate the membrane scaling, but the desalination performance would be impaired to some extents with feed velocity exceeding 0.4 m s^{-1} . The increasing feed temperature could significantly increase the membrane flux, but membrane scaling was accelerated resulting in a lower recovery.

1. Introduction

With growing application of reverse osmosis (RO) for seawater desalination, there exists a high quantity of RO brine with high salinity [1]. Discharging the RO brine directly into offshore water may result in serious coastal environment pollution and land salinization due to the high salinity of RO brine and several chemical agents added to pretreat seawater prior to RO [2–4]. In the meantime, discharging RO brine may also cause a waste of water resources leading to a higher burden of associated facilities [5]. Therefore, to treat and to recycle RO brine is of great importance for seawater desalination industry.

A variety of technologies were investigated to treat RO brine, including pressure-driven membrane processes, current-driven membrane processes and thermal-driven processes [6,7]. The pressure-driven membrane processes, such as RO and nanofiltration, were able to produce high quality water. However, a much bigger energy demand resulting from concentration polarization and membrane fouling constricts the application of the aforementioned pressure driven processes. The current-driven membrane processes like electro-osmosis have the same advantages, but the energy consumption is proportional to the brine salinity and the rate of salt removal [8]. Hence, both pressure-driven and current-driven membrane processes are not economically

feasible in treating high-salinity RO brine [6]. In contrast, thermal-driven processes possess a great potential in the brine treatment due to their insensitivity to the high salinity [9]. Solar evaporation is commonly used for the brine disposal at a low cost [10,11], but the large land demand for evaporation pond restricts its application, especially in some locations with slow evaporation rates [10]. Besides, leakage of evaporation ponds may pose a high contamination potential to groundwater [12].

Membrane distillation (MD) is a thermal-driven separation process combined with membrane technology. In the MD process, volatile compounds evaporate on the feed side and are collected on the other side of membrane. According to the steam collection form, MD can mainly be classified into direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweep gas membrane distillation (SGMD) and vacuum membrane distillation (VMD) [13]. DCMD, which has the simplest configuration, is widely used in the scientific investigation of MD [14]. Compared to traditional evaporation processes, MD is able to be operated within a relatively moderate temperature range (40°C to 80°C), which makes it far more compatible with low-grade waste heat sources or renewable energy [13]. In addition, membrane modules can be arranged compactly due to small vapor space demand, considerably reducing the footprint of desalination

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plants [15]. Because the vapor pressure of feed is not as strongly dependent as osmotic pressure, a relatively high recovery rate can be expected in MD process. Therefore, MD becomes attractive for RO brine treatment in recent years. Qu et al. [16] concentrated the primary RO brine by 40 times using DCMD, and achieved a water recovery rate as high as 98.8%. Mericq et al. [5] have performed a study on the seawater reverse osmosis brine treatment using VMD, and an overall recovery factor of 89% was achieved by coupling RO and VMD via a simulation analysis.

Despite great desalination performance presented in literature, MD has not been extensively applied for industrial desalination [17]. It is considered that low energy efficiency and lack of commercially available and high performance membranes constrict the commercialization of MD [18]. However, as the development of multi-stage MD, energy efficiency or gained output ratio (GOR) in MD was comparable to other thermal-driven technologies (multistage flash, MSF; multiple effect evaporation) [19]. In addition, low-grade thermal energy and renewable energy can be used in MD, which will effectively decrease the operation cost of energy (close to or lower than RO) [19,20]. With regard to commercial MD membranes, plenty of excellent and high-performance membranes are fabricated in the lab recently [21–23]. Therefore, MD is promising for the industrial application in the near future. Similar to other membrane process, membrane scaling is another factor substantially constricting the application of MD. Given the growing number of studies on the scaling in other membrane processes (such as NF, RO), scaling of sparingly soluble salt is yet to be fully understood in a high-concentration and high-temperature environment adjacent to hydrophobic MD membranes [17,24]. The deposition of pollutants and crystals on the membrane surface may lead to severe wetting, resulting in permeate deterioration [25,26]. Moreover, scaling will also affect heat conduction and thermal performance, but it is hardly analyzed. Therefore, it is significant to investigate the scaling and fouling phenomena in the MD process.

In this study, DCMD was performed to treat a simulated RO brine containing a small amount of sparingly soluble salt (calcium sulfate). Pure water was used to investigate the effects of feed velocity and temperature on clean water flux of the MD membrane. Heat transport analysis was conducted to study the temperature polarization and thermal performance in the MD module. RO brine treatment using DCMD was performed to investigate the efficiency of MD at a series of feed temperatures and velocities. Membrane fouling and wetting were monitored during the whole process. Membrane scaling was characterized with respect to morphology and reversibility.

2. Materials and methods

2.1. Membranes and feed water

A hydrophobic PVDF membrane (IPVH00010), which was purchased from Millipore Corp (Millipore, MA, USA), was used in this study. The PVDF membranes were characterized with respect to contact angle, porosity, maximum pore size and thickness beforehand. The chemical agents, including sodium chloride (NaCl), potassium chloride (KCl), magnesium sulfate ($MgSO_4$), magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$) and calcium chloride ($CaCl_2$), were purchased from Sinopharm Chemical Reagent Corp (Beijing, China). The simulated RO brine was prepared in lab containing 32.61 g/L NaCl, 1.03 g/L KCl, 6.96 g/L $MgCl_2 \cdot 6H_2O$, 4.50 g/L $MgSO_4$ and 4.50 g/L $CaCl_2$ according to the composition of RO brine given by Ge et al. [27]. The mixed solution was filtered by a 0.45 μm microfiltration membrane (Taoyuan, China) subsequently. The conductivity of the feed was determined as $62 \pm 1 \text{ mS cm}^{-1}$.

2.2. DCMD experiment

The RO brine treatment by DCMD was conducted in a bench-scale

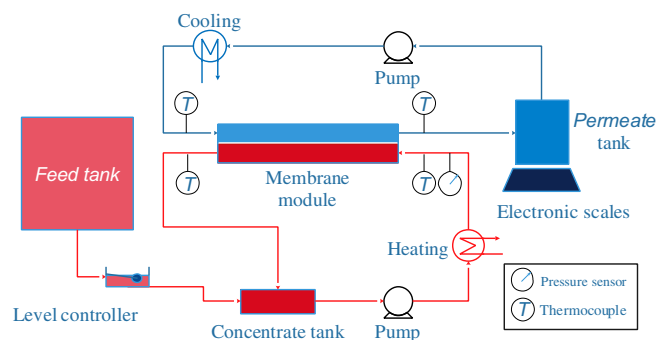


Fig. 1. DCMD experimental setup.

self-manufactured experimental setup of which the schematic diagram is shown in Fig. 1. The DCMD system was comprised of a feed circulation system, a permeate circulation system and a membrane module. In the feed circulation system, 200 mL feed was added to the concentration tank beforehand, and raw water was supplemented by a feed tank to maintain a constant water volume in the feed circulation. Feed was continuously pumped to go through a heat exchanger and the membrane module before returning to the concentration tank using a peristaltic pump (WT3000-1JB, Longer, China). The heat exchanger was adopted to maintain the water temperature of feed at $50 \pm 1^\circ C$. In the permeate circulation system, 200 mL of Milli-Q deionized water was initially added to serve as a cold medium. The permeate was circulated through a cooling unit and the membrane module using a peristaltic pump. A thermostat bath (DC-0510, Scientz, Ningbo, China) was applied to maintain the permeate temperature at $20 \pm 1^\circ C$.

A flat sheet MD membrane with an effective area 25 cm^2 was placed inside the membrane cell with a plastic mesh used as the support. The feed and permeate in both sides of the MD membrane were circulated in the opposite direction. The permeate velocity was fixed at 0.25 m s^{-1} , and the feed velocity varied from 0.05 to 0.4 m s^{-1} . Inside the cell, the feed evaporated in the feed side, and then the vapor penetrated through membrane pores due to the vapor pressure gradient induced by the temperature difference. Finally, the vapor condensed on the permeate side. The permeate tank was placed on an electronic balance connected to a computer with weighing data automatically logged every 5 min. To predict pore wetting, a conductivity meter (Seven Compact S230, Mettler Toledo, Switzerland), which was connected to the computer, was installed to monitor the permeate conductivity on line with an electrode (InLab 741, Mettler Toledo, Switzerland). In addition, the hydraulic pressure of feed at the cell inlet was also monitored using a pressure transducer (PTP708, Tuopo Electric, Foshan, China). Because the circulation flow rate was maintained constant, an increase in feed pressure could manifest the membrane scaling which would reduce the cross-section area for the circulated water flow.

2.3. Membrane characterization

Membrane was pre-dried in the vacuum drying oven ($70^\circ C$ and -0.08 MPa) for one day before characterization. Contact angle was measured by an optical contact angle meter (JYSP-180, Jinshengxin Co., Ltd., China) to evaluate the hydrophobicity of the MD membrane. A square piece of the MD membrane ($0.5 \text{ cm} \times 0.5 \text{ cm}$) was fixed on a slide glass which was placed on the sample stage of the device. $3 \mu L$ water was dripped on the membrane surface at room temperature ($25^\circ C$) using a microsyringe. The images of water drop were captured in 3 s and then the contact angle could be obtained using a three-point method. The measurement was repeated for 6 times.

Liquid entry pressure of water (LEP_w) was used to evaluate the tolerance of MD membrane to pore wetting. A piece of disk membrane (diameter 25 mm) was fixed on a self-made filtration cell (as shown in Fig. S1) to determine the LEP_w . Detailed instruction of measurement

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