



# Treatment of high salinity brines by direct contact membrane distillation: Effect of membrane characteristics and salinity



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## HIGHLIGHTS

- Four PTFE membranes were examined in treating highly concentrated salt solutions.
- Active layer of membrane is crucial to mass transfer coefficient and system thermal efficiency.
- Flux decreases significantly as salt concentration increases to more than 2.0 M.
- High salinity KCl, NaCl and MgCl<sub>2</sub> solutions ( $\geq 2.0$  M) shows distinct trends over flux decline.

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## ABSTRACT

Direct contact membrane distillation (DCMD) is one of the attractive technologies for high salinity brine treatment. In this study, four polytetrafluoroethylene (PTFE) membranes were examined in treating highly concentrated salt solutions. Results showed that non-supported membranes generally have a higher overall mass transfer coefficient but porosity seems to be the most important parameter controlling membrane flux and thermal efficiency. Supported membranes with large thickness had relatively higher thermal efficiency than small thickness. This can be attributed to their reduced heat loss through heat condition. In addition, KCl, NaCl and MgCl<sub>2</sub> solutions showed distinct trends over flux decline at high salt concentrations ( $\geq 2.0$  M). The difference in flux was largely due to the discrepancy in water activities of these solutions (KCl > NaCl > MgCl<sub>2</sub>). However, the effect of viscosity on permeate flux could not be neglected for MgCl<sub>2</sub> at high salt concentrations as the suddenly increased viscosity could lead to serious temperature polarization. This study indicates that membrane distillation is a promising technology for high salinity brine treatment.

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## 1. Introduction

Most Chinese potash is produced from salt lake brines, and evaporation ponds are often utilized for concentration of saline water and precipitation of minerals, which are then further processed in chemical plants (Wang et al., 2014). In the saline water of the evaporation ponds, NaCl concentration is often greater than or equal to 2.0 M, and it also has a large amount of other salts like KCl and MgCl<sub>2</sub> (Hickenbottom and Cath, 2014; Wang et al., 2014). However, evaporation ponds are time and energy intensive, and when used, large volumes of valuable water are lost to the

atmosphere. Replacing the traditional evaporation ponds with desalination processes could minimize land use and also increase water recovery from the salt lake brine (Hickenbottom and Cath, 2014). Nevertheless, the desalination of this high salinity brine has been regarded as a challenge in desalination industry (Greenlee et al., 2009; Adham et al., 2013; Hickenbottom and Cath, 2014). It is often hampered by concentration polarization and fouling (scaling) issues associated with high salinity.

Membrane distillation (MD) is a separation process that combines membrane and evaporation technology (Alkudhiri et al., 2012). In MD process, the hydrophobic membrane itself acts as a barrier to hold the liquid/vapor interfaces at the entrance of the pores, in which only vapor or volatile substances is able to pass through the membrane (Lawson and Lloyd, 1997; El-Bourawi et al., 2006). The driving force of MD is vapor pressure difference across the hydrophobic membrane, which is quite different from

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conventional membrane processes (i.e., nanofiltration, reverse osmosis) (Wang and Chung, 2012). One of the most important advantages of MD is the ability to treat high salinity brines, as it is less sensitive to membrane fouling and feed salinity (Adham et al., 2013; Hickenbottom and Cath, 2014). It has also been reported that salt rejection was independent on feed salinity in treating high salinity brines (Safavi and Mohammadi, 2009). In addition, incorporating with crystallization unit, MD can also recover crystal valuable products from high salinity brines (Tun et al., 2005; Edwie and Chung, 2012).

However, it should be noted that the high salinity may have adverse effects even for MD (Yun et al., 2006; Edwie and Chung, 2012), as permeate flux could decrease at high salinities due to the change of water activity and viscosity (Hickenbottom and Cath, 2014; Guan et al., 2015). In the literature, Song et al. (2007) reported a hydrophobic polypropylene (PP) membrane with a porous fluorosilicone coating on the outside surface in DCMD-based desalination. A small flux reduction was observed when the NaCl concentration increases to 10 wt% NaCl and an extended 5-d run did not show any pore wetting. Edwie and Chung (2012) have designed and investigated three types of membrane for water and salt recovery from high salinity sodium chloride (24 wt% NaCl) via DCMD and crystallization. They found that the single-layer PVDF membrane exhibited a superior wetting resistance as compared to dual-layer membranes. Lee et al. (2011) developed a countercurrent cascade of the novel cross-flow DCMD system integrating with a heat exchange device for desalination of artificial seawater. By the time that the artificial sea water was concentrated from 3 times (10.5 wt% NaCl) to 8 times (26 wt% NaCl), the water vapor production rate had declined from 86% to 65% of the original steady-state production rate. It is obvious that the increased concentration of the solutes will decrease the vapor pressure of water, which drives the water vapor flux. These studies are very helpful for development and optimization of membrane or membrane modules specified for high salinity brine desalination. However, it remains one of the major challenges to source out a high efficient but low-cost membrane in the market. In this regard, the prime objective of this study was to examine the performance of four different commercial PTFE membranes for high salinity brine treatment ( $\geq 2.0$  M). The effect of membrane characteristics such as porosity, membrane thickness and with or without support layer on mass transfer coefficient and thermal efficiency of DCMD were investigated. In addition, the effect of salt types (i.e. KCl, NaCl and  $MgCl_2$ ) and salt concentration (0–4.0 M) on DCMD performance was also studied. It is believed that this work would help to facilitate the design and optimization of DCMD process for high salinity brine treatment.

## 2. Materials and methods

### 2.1. Experimental setup

Fig. 1 shows a schematic diagram of the experimental setup. The high salinity aqueous solutions were stored in a 2 L three-neck flask. The flask was submerged in a thermostatic water-circulator bath (HH-501A, Jiangsu China,  $\pm 0.1$  °C) to maintain the desired feed temperature. The deionized water was used as the cooling agent in the product loop, and the temperature was controlled through a chiller. Tap water was used as coolant of the chiller and chilled by a precise low-temperature thermostat bath (DS-2006, Ningbo, China,  $\pm 0.1$  °C). The product overflowed from the permeate reservoir (2 L) to a conical flask, which was continuously weighed by an electronic balance (DJ-1000J, Shanghai Zhongchen Co., Ltd., China). The flat-sheet membrane module used in this study was made by nylon fiberboard (50 mm thickness). Two fiberboards with eight engraved flow channels acted as the feed and permeate

semi-cells respectively. Each channel is 90 mm  $\times$  3 mm  $\times$  2 mm (L  $\times$  W  $\times$  H), with the total effective membrane area of 14.4 cm<sup>2</sup>. In all experiments, the membrane module was maintained in a horizontal position. The membranes used in the experiments were PTFE hydrophobic membranes with different characteristics, as listed in Table 1.

The circulation of the feed and permeate was in countercurrent direction, and the flow rates of both side were maintained identical all the time. In order to lower the resistance to vapor flow through the membrane due to the presence of trapped air in the pore (Cath et al., 2004), the permeate stream was pumped out of the membrane cell in this work, and a vacuum of 6 kPa on the permeate side was induced in the experiments. The permeate conductivity was continuously monitored by a conductivity meter (Seven Multi, Mettler Toledo, American). The inlet and outlet temperature of feed ( $T_{fi}$  and  $T_{fo}$ ) and permeate ( $T_{pi}$  and  $T_{po}$ ) were continuously measured using thermometers on site. Temperatures, pressures, and flow rates were continuously monitored, and product weight and permeate conductivity were recorded every 10 min when the system reached a steady state. The experiment was stopped when flux was unchanged for at least 1 h.

A 2.0 M NaCl solution was used to represent the high salinity brine in the experiments expect for the study of different salt types and salt concentrations. Sodium chloride (NaCl), Potassium chloride (KCl), and Magnesium chloride hexahydrate ( $MgCl_2 \cdot 6H_2O$ ) were analytical reagents purchased from Tianjin Bodi Chemical Co., Ltd, and deionized water was used to prepare the feed solutions.

### 2.2. Operational parameters

In all the experiments, the cooling water in the permeate reservoir was maintained at 15 °C. The inlet temperature  $T_{fi}$  of feed solution was varied from 50 °C to 70 °C at the increment of 5 °C to study the effect of feed inlet temperature on thermal efficiency. Otherwise it would be maintained at 55 °C. Similarly, the recirculation rate was in the range of 0.2 m s<sup>-1</sup> to 0.5 m s<sup>-1</sup> for testing different recirculation flow rates, otherwise it would be maintained at 0.3 m s<sup>-1</sup>. NaCl, KCl and  $MgCl_2$  were used to investigate the effect of salt type on permeate flux. The concentration of these salts varied from 0 M to 4.0 M at the increment of 0.5 M to study the influence of salt concentration on permeate flux, in which the feed inlet temperature, cooling reservoir temperature and recirculation flow rate were maintained at 50 °C, 15 °C and 0.3 m s<sup>-1</sup> respectively.

### 2.3. Analytical methods

The morphology of active layer and cross section of the membranes were examined by a SEM (JSM-7001F, JEOL, Japan). The membranes were first frozen in liquid nitrogen for a few minutes and then cut into pieces with a blade in this state to prevent deformation. The samples was coated with platinum by an auto fine coater (JEOL, JFC-1600) before the examination. Membrane thickness and active layer thickness were determined from the obtained SEM cross-section images, and measured 3 times from different sections of the membrane, and the average was reported. The porosity of membranes was determined by mass difference (Zhang et al., 2010). In all the experiments, the salt rejections were greater than 99.5%, therefore the salt rejections were not included in the discussion.

### 2.4. Determination of overall mass transfer coefficient and system thermal efficiency

It is believed that the mass transfer rate or water flux in membrane distillation is proportional to vapor pressure difference across the membrane (Schofield et al., 1990).

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