



Membrane fouling and wetting in a DCMD process for RO brine concentration



Ju Ge^a, Yuelian Peng^{a,*}, Zhehao Li^b, Ping Chen^c, Shaobin Wang^d

^a Department of Chemistry and Chemical Engineering, College of Environmental and Energy Engineering, Beijing University of Technology, Beijing 100124, PR China

^b Changchun Gold Research Institute, 130012, PR China

^c The Research Institute of Environmental Protection, North China Pharmaceutical Group Corporation, 050015, PR China

^d Department of Chemical Engineering, Curtin University of Technology, GPO Box U1987, Perth, WA 6845, Australia

HIGHLIGHTS

- Membrane wetting was more significant at the higher feed temperature.
- Membrane fouling was exacerbated after the concentration factor reached 3.5.
- Shape of CaSO₄ crystals formed changed at different feed temperatures.
- The permeate was lower in Mg²⁺ and Ca²⁺ than Na⁺ and K⁺.

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ABSTRACT

Membrane wetting and fouling were studied using hollow-fiber polyvinylidene fluoride (PVDF) membranes to concentrate reverse osmosis (RO) brine in a direct contact membrane distillation (DCMD) process. The effect of the operating conditions, such as the feed temperature, the flow velocity and the feed solution concentration, was investigated. Membrane wetting was more significant at high feed temperatures, and the salts in the feed promoted membrane wetting; membrane wetting resulted in a decrease in the flux and salt rejection. A theoretical model was developed to simulate the DCMD process, and the results of the model were analyzed. The concentration factor significantly affected the flux and the electrical conductivity of the distillate. When the concentration factor (CF) was less than 3.5, CaSO₄ did not crystallize because of high ionic strength and higher solubility of CaSO₄ at the membrane surface than in the bulk solution; beyond CF of 3.5, membrane fouling was exacerbated because of CaSO₄ crystallization. Membrane fouling was more significant at the higher of the two temperatures investigated for long-time DCMD operation: square CaSO₄ crystals formed at 77 °C, whereas snowflakes formed at 55 °C. The permeate was lower in the divalent cations, Mg²⁺ and Ca²⁺, than in the monovalent cations, Na⁺ and K⁺.

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1. Introduction

Each year, large quantities of reverse osmosis (RO) brine discharges cause a significant loss of water resources and create disposal challenges. Thus, RO brine minimization is essential in many countries to alleviate environmental problems. Several potential methods for RO brine minimization have been explored [1,2]. Membrane distillation (MD) is a promising desalination technology for water recovery from high salinity solutions, such as RO brine, and is based on the thermally-driven transport of vapor through the pores of a hydrophobic microporous membrane. Mass and heat transfer occur simultaneously in MD [3], which is widely used because it is not limited by concentration polarization and can be used to concentrate RO brine.

However, as in all other membrane processes, very significant bottlenecks occur in MD because the permeating flux declines with time. The dominant causes of the bottleneck are membrane wetting and the adherence of scale onto the membrane surface [4]. Capillary condensation inside the membrane pores results in membrane wetting [5]. Certain salts, such as CaSO₄ and CaCO₃, have been reported to form deposits on the membrane surface or in the membrane pores, resulting in membrane wetting [2,4,6]. Ding studied the effects of a surfactant (sodium dodecyl benzene sulfonate) and an organic solvent (limonene) on membrane wetting [7]. However, few studies have been conducted thus far on membrane wetting and fouling when RO brine with a high CaSO₄ concentration is treated via MD, although it is very important to explore this technology for RO brine reduction.

Two types of scale form in RO brine, CaCO₃ and CaSO₄. Compared to CaCO₃ fouling, it is more difficult to remove CaSO₄ fouling on a membrane surface, and the removal process requires more oversight. CaCO₃ is an insoluble salt ($K_{sp}(25\text{ °C}) = 4.8 \times 10^{-9}$); CaCO₃ precipitate

* Corresponding author. Tel.: +86 10 67391090; fax: +86 10 67391983.
E-mail address: pyl@bjut.edu.cn (Y. Peng).

Nomenclature

Symbols

| | |
|------------------|--|
| C | membrane distillation coefficient ($\text{kg/m}^2 \text{ s Pa}$) |
| C_d | specific heat capacity (J/kg K) |
| C_K | mass transport coefficients for Knudson diffusion (m/s Pa) |
| C_M | mass transport coefficients for molecular diffusion (m/s Pa) |
| D | diffusivity of vapor in air (m^2/s) |
| $d_{(h)}$ | hydraulic diameter of the shell side or diameter of the lumen side (m) |
| ΔH | latent heat of water at the feed/membrane interface (J/kg) |
| L | effective length of hollow fiber membrane (m) |
| M | molecular weight of water (kg/mol) |
| N | vapor flux ($\text{kg/m}^2 \text{ s}$) |
| P_0 | partial pressure of vapor at the feed/membrane interface (Pa) |
| P_a | air pressure (Pa) |
| $P_{d,m}$ | vapor pressure at the membrane/distillation interface (Pa) |
| $P_{f,m}$ | vapor pressure at the feed/membrane interface (Pa) |
| P_m | average vapor pressure in the membrane (Pa) |
| ΔP | vapor pressure difference |
| Q_f | heat transfer from the feed to the membrane (W) |
| Q_d | heat transfer from the membrane to the distillate (W) |
| Q_m | heat transfer across the membrane (W) |
| R | gas constant (J/mol K) |
| R_i | inner radius (m) |
| R_o | outer diameter (m) |
| r | local radius (m) |
| r_{max} | maximum pore radius (m) |
| r_p | radius of membrane pore (m) |
| T | local temperature ($^{\circ}\text{C}$) |
| T_d | distillate temperature ($^{\circ}\text{C}$) |
| $T_{d,m}$ | membrane temperature at distillation side |
| T_f | feed temperature ($^{\circ}\text{C}$) |
| $T_{f,m}$ | membrane temperature at feed side |
| T_m | membrane temperature ($^{\circ}\text{C}$) |
| v | velocity (m/s) |
| v_f and v_d | velocities through the shell side and the lumen side, respectively (m/s) |
| x | molar fraction of ions in feed |
| Nu | Nusselt number |
| Pr | Prandtl number |
| Re | Reynolds number |
| LEP_w | feed liquid entry pressure (Pa) |
| TPC | temperature polarization coefficient |

Greek letters

| | |
|---------------------------|---|
| α | convective heat transfer coefficient ($\text{W/m}^2 \text{ K}$) |
| α_f and α_d | convective heat transfer coefficients of feed and distillate, respectively ($\text{W/m}^2 \text{ K}$) |
| δ | membrane thickness (m) |
| ε | porosity (%) |
| μ | gas viscosity (kg/m s) |
| θ | liquid/solid contact angle ($^{\circ}$) |
| ρ | density (kg/m^3) |
| λ_m | membrane thermal conductivity (W/mK) |
| ρ | density of water (kg/m^3) |
| τ | membrane tortuosity |

Subscripts

| | |
|------|----------------------|
| a | air |
| d | distillate |
| f | feed |
| h | hydraulic |
| K | Knudson |
| M | molecular |
| o | outside |
| i | inside |
| p | pore |
| d, m | distillate, membrane |
| f, m | feed, membrane |
| max | maximum |

usually adheres onto the vessel wall. However, the solubility of CaSO_4 is very low ($K_{sp}(25^{\circ}\text{C}) = 2.5 \times 10^{-5}$): CaSO_4 crystal nucleus form, and then they coalesce to form large CaSO_4 crystals that are suspended in the solution but do not adhere to the vessel wall like the CaCO_3 precipitate [8]. When DCMD was used to concentrate a saturated CaSO_4 solution, CaSO_4 precipitated on the membrane surface and penetrated into the membrane pores, leading to an increase in the electrical conductivity of the distillate from a few to several thousands of $\mu\text{S/cm}$ after several hours of operation [9]. Nghiem [6] used CaCO_3 (1000 mg/L) and CaSO_4 (2000 mg/L) feed solutions and unexpectedly found that CaCO_3 did not cause any discernible decline in the flux, whereas CaSO_4 produced a huge flux decline of approximately 92% after 40 h of operation. Thus, CaSO_4 fouling has more significant repercussions than CaCO_3 fouling.

The induction time appears to be a key parameter that governs the crystallization of CaSO_4 on the membrane surface and has been well-correlated with the super saturation concentration of the CaSO_4 solution. [10,11]. A low feed temperature may prolong the induction time for CaSO_4 crystallization in DCMD. The induction time was approximately 5 h at 60°C and increased to 30 h at 40°C : the flux declined by more than 90% following the induction period when a 2-g/L CaSO_4 aqueous solution was used as the feed in DCMD [6].

Despite the growing number of studies on MD, it remains unclear how CaSO_4 affects the distillate flux during the MD process. Various mechanisms for CaSO_4 crystallization in NF have been reported; however, MD and NF differ because of temperature polarization in MD, and the temperature-dependence of the solubility product and the CaSO_4 crystal structure imply that the CaSO_4 crystallization mechanism should be different in MD compared to NF. Note that studies on membrane scaling behavior and using highly concentrated RO brine as a feed solution in MD are still very scarce. In this study, we investigated how the operation parameters (i.e., the feed temperature, the feed flow rate and the concentration of salts in the feed) affect membrane wetting. A mathematical model was formulated for a DCMD process with a hollow-fiber membrane to explain the variation in the flux. To study how CaSO_4 fouling affects the flux and the electrical conductivity of the distillate in RO brine concentration, the concentration factor and feed temperature were varied in a continuous concentration experiment.

2. Theory

A schematic of the heat transfer in a single-layer hydrophobic hollow-fiber membrane is shown in Fig. 1, where the heat transfer occurs through the three sections shown below [12].

(1) Feed side: heat convection is the dominant mode of heat transfer at the feed/membrane interfaces and can be described by the following equation:

$$dQ_f = 2\pi R_i \alpha_f (T_f - T_{f,m}) dL \quad (1)$$

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