



Direct contact membrane distillation: Capability to treat hyper-saline solution



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HIGHLIGHTS

- Study the influence of operating conditions on mass transfer
- Investigate the temperature and the concentration polarization phenomena
- Discuss the permeate flux decline due to salt crystallization on membrane surface
- Supervise the permeability and the scaling of the membrane

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ABSTRACT

In this paper, we focused our work on the direct contact membrane distillation (DCMD) capability to treat hyper-saline solution. The governing operative model for mass transfer was investigated. The measured flux has been well predicted by the Knudsen-molecular mechanism model. The effects on the DCMD flux of polarization phenomena TP and CP were underlined. The optimum operating parameters were defined: the hot and cold stream temperatures were set, respectively at 59 and 20 °C, and the feed and permeate velocities were fixed both to 0.046 m s⁻¹. With regard to membrane performance to treat the reverse osmosis brine, a long-term experiment was carried out under the optimal experimental conditions. The increase in feed RO brine concentration provoked a noticeably decrease in flux from 8.43 to 4.06 kg m⁻² h⁻¹. The RO brine experiments proved that the DCMD process was capable to concentrate the solution till to reach concentration factor (CF) further than four times, which corresponded to the super-saturation of saline solution. Based on the characterization methods, the occurrence of the membrane wetting and scaling was shown and interpreted. These extreme phenomena promote the salt crystallization on the feed side of the membrane. The onset crystallization phenomenon starts when the permeate decreases so fast. Their sudden decline was about 90% for a working period of 20 h.

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1. Introduction

To cut with water scarcity many desalination plants have been installed worldwide to treat brackish water as well as seawater. Their production in fresh water was close to 90 million m³ per day in the year of 2012 [1]. This water is used to satisfy the needs of population, industry and agriculture. The desalination units use various techniques including thermal and membrane processes. Reverse osmosis (RO) and multi-stage flash (MSF) are the most used techniques overall the world [2]. RO has been the most investigated during the past 30 years [3]. A 61.1% of the worldwide capacity installed, of seawater and brackish water desalination plants, was attributed to RO.

Drioli et al. [4] estimated the optimum recovery ratio for SWRO plant to around 45–60%. The balanced part of feed stream will be discharged as brine at about 85 g L⁻¹ [1]. The generation of this concentrate effluent is one of the major RO problems. According to previous studies [5–7] done to evaluate its environmental effect, this by-product threatens hugely the ecosystem equilibrium. Thus, finding out a viable method managing RO concentrate becomes more and more challenging.

The best management is to further concentrate brines until attaining solid salt formation. In this context, classical and emerging technologies including thermal evaporation, electro-dialysis, two step RO and sequences of integrated processes were investigated [8–10]. In spite of their efficiency, these processes have significant drawbacks in term of: high investment costs, high energy consumption, needs of pretreatment stages, polarization phenomena, scaling problems, brine limited concentration Unlike, membrane distillation (MD), a promising alternative technology integrating both thermal distillation and membrane processes, doesn't suffer strongly from those limits, thanks to its ability to achieve high

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rejection of non-volatile solute and to operate at low operating pressure and temperature [11]. MD is a thermally driven process [12]. A micro-porous hydrophobic membrane [13,14] is used in the process as a physical support taken in sandwich between warm and cold solutions. The cooler chamber called the permeate side, that contains either a liquid/gas to be a matrix for vapor condensation. The membrane materializes the liquid–vapor equilibrium [15]. MD is a non-isothermal technology [16]. Thus, MD relies on temperature difference between the feed and the permeate sides of the porous membrane to create vapor pressure difference. This gradient is the driving force for this process [17,18]. The separation process taken places in three steps: the evaporation of volatile component in the feed stream, then, the migration of vapor molecules through the membrane pores from the high to the low vapor pressure side i.e., from the feed side to the permeate side, finally, the condensation of those vapor molecules which depend on the configuration of the process.

MD process admits four configurations: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), vacuum membrane distillation (VMD) and sweep gas membrane distillation (SGMD) [19]. Each one has its advantages and disadvantages and can be used in different fields. The DCMD process has been frequently investigated, for many reasons, among them, aqueous solution dewatering capacity and low flux sensitivity toward the processed feed concentration. Previous researches have been exploited these advantages. Indeed, fruit juice [20], proteins and biological solutes [21], phenolic compound [22], skim milk and whey [23], saline water from meat processing industry [24], and oil emulsions [25] have been successfully concentrated by means of DCMD units.

In the aim to crystallize salt content in RO brine discharge, we have to concentrate the RO discharge to achieve the super-saturation level which is the driving force for salt crystallization.

To achieve our objective, we start by studying the capability of DCMD to treat solution close to saturation. We proceed by the:

- Development of the predicting model for mass and heat transfer through a hydrophobic membrane applied for a hyper-saline solution.
- Validating the model by the experiments on lab-scale cell.
- Studying the effect of operating parameters on the DCMD performance.
- Determining the optimum operation parameters.
- Applying the DCMD, at its optimum operating parameters, for synthetic RO brine.
- Monitoring the permeability and the scaling of the membrane.

2. DCMD heat and mass transfer

2.1. Heat transfer

In the DCMD there are 3 regions, the feed, the membrane and the permeate region. Each one has a specific flux of heat transfer involved.

From the heated solution to the membrane surface transfer and across the feed thermal boundary layer which is the feed region, is characterized by the presence of the convective heat transfer.

Inside the membrane region, the heat is transferred by the combination of two ways; there is the conductive flux together with the heat caused by the vapor flow through the membrane.

The third region is characterized by a convective heat flux from the cold membrane surface to the cold solution.

In literature [26], the illustration of the three mechanisms of heat transfer in the feed side, through the membrane and the permeate side is described by Eqs. (1), (2) and (3).

$$Q_f = h_f(T_f - T_{fm}) \quad (1)$$

$$Q_m = Q_v + Q_c = J\Delta H_v + h_m(T_{fm} - T_{pm}) \quad (2)$$

$$Q_p = h_p(T_p - T_{pm}) \quad (3)$$

where T_f , T_{fm} , T_p , T_{pm} , ΔH_v , h_f , h_p , h_m and J are, respectively, the bulk feed, the surface feed, the bulk permeate, the surface permeate temperatures, the latent heat of vaporization, the feed side, the permeate side, the membrane heat transfer coefficients and the permeate flux.

$$h_m \text{ is given by: } h_m = \frac{k_m}{\delta} \quad (4)$$

δ is the nominal membrane thickness (m) and k_m is the effective membrane conductivity ($\text{Wm}^{-1} \text{K}^{-1}$) estimated using the vapor and solid phase thermal conductivities k_v and k_s .

$$k_m = (1 - \varepsilon)k_s + \varepsilon k_v \quad (5)$$

where m , s , and v , refer to the membrane, solid and vapor phases and ε is the membrane porosity.

k_m is the assuming constant over the range of temperatures considered with steady-state conditions, heat flux is constant in all regions, this means that: $Q_f = Q_m = Q_p = Q$.

The ability to calculate or to measure experimentally the membrane surface temperatures of the feed and permeate sides T_{fm} and T_{pm} is not allowed. But, thanks to the energy balance and using Eqs. (1), (2) and (3), a mathematical iterative model defines the two following Eqs. (6) and (7) to estimate, respectively, feed membrane surface temperature and permeate membrane surface temperature [27].

$$T_{fm} = \frac{\frac{k_m}{\delta} \left(T_f + \left(\frac{h_f}{h_p} \right) T_p \right) + h_f T_f - J\Delta H_v}{\frac{k_m}{\delta} + h_f + \frac{k_m h_f}{\delta h_p}} \quad (6)$$

$$T_{pm} = \frac{\frac{k_m}{\delta} \left(T_p + \left(\frac{h_p}{h_f} \right) T_f \right) + h_p T_p - J\Delta H_v}{\frac{k_m}{\delta} + h_p + \frac{k_m h_p}{\delta h_f}} \quad (7)$$

h_f and h_p which depend on the working conditions are calculated from this equation.

$$h_i = \frac{Nu_i k_i}{d_h}, \quad i = f, p \quad (8)$$

where k_i is the thermal conductivity, d_h is the hydraulic diameter and Nu is the Nusselt number. According to the literatures [27–29], there are many Nusselt number relations for laminar and turbulent flows. Its empirical form is:

$$Nu = \text{Constant } Re^a Pr^b \quad (9)$$

where Re and Pr are the Reynolds number and the Prandtl number.

The well-known used equations for the laminar and turbulent flow are respectively [30]:

$$Nu = 0.13 Re^{0.64} Pr^{0.38}, \quad Re < 2100 \quad (10)$$

$$Nu = 0.023 Re^{0.8} Pr^{0.33}, \quad Re > 2100. \quad (11)$$

Re presents the Reynolds number $Re = \frac{\rho v d_h}{\mu}$ Pr presents the Prandtl number $Pr = \frac{C_p \mu}{k_m}$ where ρ , v , μ and C_p are, respectively, the density, the average velocity, the viscosity and the specific heat.

2.2. Mass transfer

The DCMD mass transfer process can be described by two steps. The volatile component crosses the concentration boundary on the feed side, afterward the porous membrane. Referring to Darcy's law the

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