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Influence of salt concentration on DCMD performance for treatment of highly concentrated NaCl, KCl, $MgCl_2$ and $MgSO_4$ solutions

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HIGHLIGHTS

- Permeate flux declines for NaCl, KCl, $MgCl_2$ and $MgSO_4$ as concentration increases.
- Permeate flux of the solution follows the order of KCl > NaCl > $MgSO_4$ > $MgCl_2$.
- The reverse flux of $MgCl_2$ results from its low water activity and high viscosity.
- Effects of circulation velocity on flux are notable for higher viscosity fluids.
- An appropriate temperature range is a must to prevent reverse flux and scaling.

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ABSTRACT

Highly concentrated NaCl, KCl, $MgCl_2$ and $MgSO_4$ solutions were treated using DCMD. The effects of salt concentration (1.0–4.0 mol/L) and circulation velocity (0.1–0.5 m/s), as well as thermodynamic and physical properties of the salt solutions on permeate flux were investigated. Results showed that the permeate fluxes decrease with increasing concentration for the four salts solutions studied, which follows the order of KCl > NaCl > $MgSO_4$ > $MgCl_2$ at the salt concentrations higher than 1.0 mol/L. The different vapor pressure depression caused by reduction of water activity was identified as the main reason behind this. However, the drastic increase of viscosity of $MgSO_4$ and $MgCl_2$ solutions at higher salt concentrations would also have a notable adverse impact on permeate flux. Under these circumstances, change of hydrodynamics, i.e. increase of circulation velocity would be a great help to improve the heat transfer and then the flux. To prevent salt from crystallizing on membrane surface in saturated conditions, the feed inlet temperature should be controlled within a certain range, and it was 40 to 50 °C, 40 to 45 °C and 25 to 35 °C for NaCl, KCl, and $MgSO_4$, respectively in this study.

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1. Introduction

With the growth of the population all over the world, water scarcity for drinking, industrial, or irrigation purposes is gradually becoming a global issue. To date, the feasible methods to increase water supply beyond what is available from the hydrological cycle are desalination of seawater/brackish water [1]. Among the varieties of approaches proposed in desalination, reverse osmosis (RO) has been regarded as one of the most promising technologies to increase the supply of drinking water. However, in most cases, the water recovery is less than 55% [1], and a large amount of brine with a higher salinity of 65,000–85,000 mg/L is generated from RO process [2]. These concentrated brines are often disposed to the lakes, rivers or oceans, which would be harmful to the surface, ground water and ecosystem [2,3]. Further treatment of concentrated brines is desired to reduce the negative impact of concentrated brines on the environment and enhance water recovery, as well as recovery of valuable inorganic elements [1,4,5].

Membrane distillation (MD) is a separation process that combines membrane and distillation technology. Due to its potential advantages, such as lower operating temperature/hydrostatic pressure (lower than

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its boiling point under pressures near atmosphere), resistance to membrane fouling, less sensitivity to feed salinity, and small volume of discharge, the MD has been recognized as one of the most effective techniques for treatment of highly concentrated brines, especially in places where waste heat, solar or geothermal sources are available [6].

Several researchers have focused on treatment of concentrated solutions with MD [7–11], and believed the flux decline with salt concentration is one of the challenges. Schofield et al. [7] found that decrease in the flux with the increase of NaCl concentration, is mainly caused by the decreases of the equilibrium vapor pressure of NaCl solution. Martínez [8] reported that a flux decrease of NaCl solution is mainly due to the decline in water activity; Apart from water activity, viscosity was another factor contributing to flux decline duing MD of sucrose solution. Safavi and Mohammadi [9] reported a method of improving MD performance through decreasing feed concentration during treatment of concentrated NaCl, $MgCl_2$ and KCl solutions with vacuum MD.

Another concern for treating highly concentration brines is the effect of polarization and/or salt crystallization on the membrane surface, leading to the failure of MD process. Yun et al.[12] have studied highly concentrated NaCl solutions in direct contact membrane distillation (DCMD). It was found that water fluxes decline sharply as the membrane surface concentration of NaCl solution reaches saturation, which was mainly attributed to the concentration polarization resistance and membrane fouling resistance. Similar results have also reported for NaCl and Na_2SO_4 solution at nearly saturated conditions by Tun et al. [13]. In their study, a rapid flux decline was observed over critical degree of saturation for both salts, which can be explained by crystal deposition and scale formation on the membrane. But Na_2SO_4 solution was able to operate at slightly higher degrees of saturation in comparison to NaCl. Edwie and Chung [14] reported that membrane flux increases with feed temperature, and found membrane scaling and wetting come severe at higher feed temperatures (60 °C and 70 °C) for a saturated NaCl solution.

Although attempts have been made on identifying the effects of high salinity on the MD performance, it should be noted that there is still lack of detail information on comparison of DCMD behavior for treating different highly concentrated salt solutions, e.g. NaCl, KCl, $MgCl_2$ and $MgSO_4$. In addition, membrane fouling and/or additional resistances associated are still not clear when the solutions are further concentrated to nearly saturated conditions. Therefore, this work aims to study the impact of different salt type and salt concentration on DCMD performance. To have an in depth understanding, the thermodynamic properties and rheological properties of NaCl, KCl, $MgCl_2$, and $MgSO_4$ solutions at different concentrations were discussed. Another objective of this work is to optimize the operating conditions of DCMD when NaCl, KCl, $MgCl_2$ and $MgSO_4$ solutions come to saturated conditions. It is expected this study would provide a further understanding of the different MD behaviors of NaCl, KCl, $MgCl_2$ and $MgSO_4$ solution during the membrane distillation of concentrated solutions.

2. Materials and methods

2.1. Experimental materials

Sodium chloride (NaCl), Potassium chloride (KCl), Magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$), and Magnesium sulfate heptahydrate ($MgSO_4 \cdot 7H_2O$) were analytical reagents and were purchased from Tianjin Bodi Chemical Co., Ltd. Solutions of NaCl, KCl, $MgCl_2$ and $MgSO_4$ were prepared by dissolving the salts into DI water. The saturated solutions of NaCl, KCl, $MgCl_2$ and $MgSO_4$ were prepared at ambient temperature as follows: a sufficient amount of salt was dissolved in deionized water and stirred overnight to achieve saturation, and then the saturated solution was stored for another 12 h and filtered with a 0.45-micron filter to remove any particles from the solutions.

A microporous commercial flat membrane was used in this work, which was provided by Sumitomo Electric Industry Ltd Corp. (Japan). The membrane consists of a thin porous polytetrafluoroethylene (PTFE) active layer and a polypropylene non-woven fabric net support layer. The main structural characteristics as specified by the supplier are 35 μm thickness of active layer, 0.22 μm nominal pore size and 82% porosity.

2.2. Experimental setup

The schematic diagram of the experimental setup is illustrated in Fig. 1. The feed solutions were in a 2 L three-neck flask submerged in a thermostatic water-circulator bath (HH-501A, Jiangsu, ±0.1 °C) to maintain the desired temperature. The permeate temperature was controlled through a precise low-temperature thermostat bath (DS-2006, Ningbo, ±0.1 °C). The water overflowed from the permeate reservoir (2 L) to a conical flask, which was continuously weighed by an electronic balance (DJ-1000J, Shanghai) and recorded every 10 min. The permeate flux was calculated after flux became constant (at least 1 h). The flat-sheet membrane module was made of nylon fiberboard (50 mm thickness). Two fiberboards with eight engraved flow channels and the flat sheet membrane are sandwiched. Each channel was 2 mm wide, 3 mm deep, and 90 mm long with the total active membrane area for mass transfer is 14.4 cm^2.

The recirculation of the fluids on both side of the membrane was in countercurrent directions controlled by two independent peristaltic pumps (WT600-2J, Baoding, China) with variable motor velocity. Temperature, pressure, and flow rate were continuously monitored. The flow rate was measured with rotometers on each side of the membrane, and the circulation velocity of the both sides was kept identical over the entire experimental period. The inlet and outlet temperature of feed ($T_{f,i}$ and $T_{f,o}$) and permeate ($T_{p,i}$ and $T_{p,o}$) streams were measured using mercury thermometer. The conductivity of the permeate reservoir was continuously monitored using a conductivity meter (SevenMulti, Mettler Toledo), and changes in conductivity were used to calculate salt rejection subsequently. In order to maintain the feed concentration, deionized water was added to feed container to replenish the permeate loss every 10 min. In all cases, deionized water was recirculated in the cold semicell from the beginning of each run.

2.3. Membrane distillation experiments

Experiments were carried out to investigate the effects of salt concentration (1.0 — 4.0 mol/L) and circulation velocity (0.1–0.5 m/s) on permeate flux for NaCl, KCl, $MgCl_2$ and $MgSO_4$ solutions. The MD performance of the nearly saturated solutions was studied over the feed inlet temperature ranged from 25 °C to 65 °C, and the permeate temperature was controlled at 20 °C for the entire experimental period. The membranes were directly taken out from the module, rinsed with ethanol absolute and stored in a desiccator before analysis. Then it was cut into pieces with a blade and coated with platinum by an auto fine coater (JEOL,JFC-1600) before the examination with Field Emission Scanning Electron Microscope (FESEM) (JSM-7001 F, JEOL).

2.4. Measurement of viscosity of the salt solutions

The viscosity measurements for 1.0 to 4.0 mol/L NaCl, KCl, $MgCl_2$ and $MgSO_4$ solutions were performed using an Ubbelohde Viscometer (JC522-1835, Beijing baiwan technology centre, China) [15]. The viscometer was inserted into the holder and placed in the constant temperature bath with the temperature set at 50 °C. Each test was repeated 3 times, and the average was recorded.

2.5. Determination of water activity in solutions

The thermodynamic property of salt solution was simulated with OLI Stream Analyzer™ (OLI Systems, Inc., Morris Plains, NJ; version 3.2) to determine the water activities of the salt solutions (i.e. NaCl,

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