



Study on vacuum membrane distillation of PP hollow fiber membranes used in concentrated seawater from low-pressure reverse osmosis

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ABSTRACT

Concentrated seawater from low-pressure reverse osmosis (RO) was treated by membrane distillation technology (MD). Hydrophobic polypropylene (PP) hollow fiber was fabricated by the melt-spinning method. The microstructure of the PP hollow fiber membrane was observed. The effects of NaCl concentration, flow rate, and temperature on water flux and rejection rate were studied. The results showed that water flux and salt rejection rate changed a little with increasing initial concentration of NaCl solution. Water flux increased greatly with the increase in feed temperature, vacuum pressure and solution flow rate. The scanning electron microscopy (SEM) result indicated that the slit-like micropore size well formed with the maximal pore size $0.05\ \mu\text{m}$ wide while $0.3\ \mu\text{m}$ long. In this work, the maximal water flux reached $7.8\ \text{L}/\text{m}^2\text{h}$, and the rejection ratio of NaCl was above 99.9%.

Keywords: Membrane distillation; Polypropylene; Hollow fiber; Distillation

1. Introduction

Membrane distillation (MD), first brought forward by Findley in the mid-1960s [1], is a new separation process which is driven by a transmembrane vapor pressure difference. MD has been regarded as an economical and efficient method for seawater distillation, and it can reach a desalting efficiency of 100%, while reverse osmosis (RO) can only reach 95–98% [2]. Compared with other membrane processes, the advantage of MD is that it can concentrate an aqueous solution with non-volatile solutes to an extremely high concentration; while RO has requirements in water

concentration because of osmotic pressure. MD can also use low-quality heat sources, for example, solar energy, geothermy, and waste heat [3].

The hydrophobic nature of the membrane prevents the penetration of the aqueous feed into the membrane, whenever the breakthrough pressure is not reached. The membrane material of MD should meet the conditions of hydrophobicity and appropriate pore size that are resistant to wetting, and the main types are polytetrafluoroethylene (PTFE), polypropylene (PP), polyethylene (PE), and polyvinylidene fluoride (PVDF).

At present, the water yield of RO technology can only reach 70%, that is, to say about 30% concentrated

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water is discharged to environment directly, which not only bring high salinity to environment, but also waste a lot of water resources. To reduce the emissions of the concentrated water of RO, and to improve water production rate, home and abroad researchers did a lot of research, but the effect is not ideal. In the recent years, more and more people pay greater attention to MD in high salinity and reuse areas. Some recent works have focused on the use of MD in concentrated seawater from low-pressure RO. MD also has a good application in other areas of film processing, for example traditional Chinese medicine and industrial wastewater treatment. Pan et al. [4] pointed out that there were many problems in MD, which need to be solved, such as film cost, intensity, hydrophobic, aging, flux, etc.

Urtiaga studied kinetics of the vacuum MD of chloroform from aqueous solution using pp hollow fiber membranes and analyzed mass transfer in laminar and turbulent state. The result indicated that only at higher Reynolds numbers within the turbulent flow regime the resistance to mass transfer in the membrane had influence on the overall mass transfer coefficient [5]. Gryta researched the influence of polypropylene membrane surface porosity on the performance of MD process, and the result indicated that a low surface porosity did not limit the possibility of surface wetting of polypropylene membranes, but hindered the scale formation inside the pores [6].

Na and Ma [7] used pp flat membrane prepared by thermally induced phase separation in seawater distillation. The average membrane pore size is $0.1\ \mu\text{m}$ measured by mercury intrusion method. With vacuum pressure 3 kPa, feed temperature is 323.15 K, and the flow rate 50 L/h, the average water flux reached $10.56\ \text{L}/\text{m}^2\text{h}$, and rejection rate reached 99.8%. Lv [8] did a detailed study of PVDF membrane used in seawater distillation, and the results of the study showed that the maximum water flux could reach $17.6\ \text{L}/\text{m}^2\text{h}$.

Microporous hydrophobic polypropylene hollow fiber membranes have had a wide application in the separation field for the past few years because of their good mechanical strength, thermal and chemical stability, good biocompatibility, large surface area and much lower prices [9].

In this work, the vacuum distillation performance was investigated considering feed temperature, concentration and pH of NaCl solution, velocity of flow and vacuum degree. The MD operation conditions on the flux and the influence of distillation rate are discussed.

2. Experimental section

2.1. The experiment equipment and materials

Polypropylene hollow fiber membrane: home-made, with contact angle about 108° and the maximal pore size $0.05\ \mu\text{m}$ wide while $0.3\ \mu\text{m}$ long. Analytical Balance: Model TG328A, Shanghai Balance Instrument Factory. Field emission scanning electron microscopy: (FESEM, Hitachi-s-4,800).

2.2. Preparation of membrane component and its parameters

U-shaped filter membrane component was tested. As shown in Fig. 1 and in Table 1, we listed the parameters of the membrane component, and pipe diameter is the diameter of the pipe in which the membrane component is installed and also the channel in which the NaCl solution flows. Porosity was tested according to the weighing method [10]; pore size was represented by electron microscopy.

2.3. MD experiment device and operation

The experimental setup included constant temperature water bath, vacuum pump, the hollow fiber membrane module, and condensing units as shown in Fig. 2.

Having added the NaCl solution, we configured in a beaker at a constant temperature water bath, while installing the membrane module. First, the water was heated in the bath to a certain extent, and then NaCl solution was put into the membrane tube with starting the magnetic pump and the cycle outside the membrane. The vacuum pump was turned on in order to pump inside the membrane to a certain vacuum with a release valve for adjustment. The water vapor permeated through the membrane from outside to inside. In the end, it reached the collection bottle. Electrical conductivity of the water was measured by the conductivity meter.

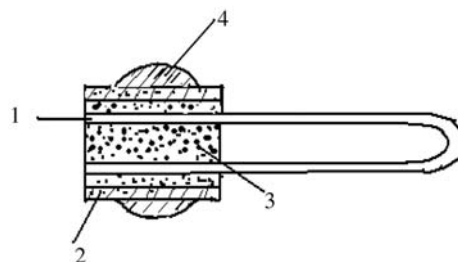


Fig. 1. U shape component of membranes (1. membrane fiber, 2. plastic pipe, 3. A/B glue, and 4. the section).

Table 1
Parameters of membrane component

Fiber number	60
Inner diameter, mm	0.028
Outside diameter, mm	0.035
Useful length, cm	12
Porosity, %	60
Pore size, μm	0.05–0.3
Pipe diameter, cm	10

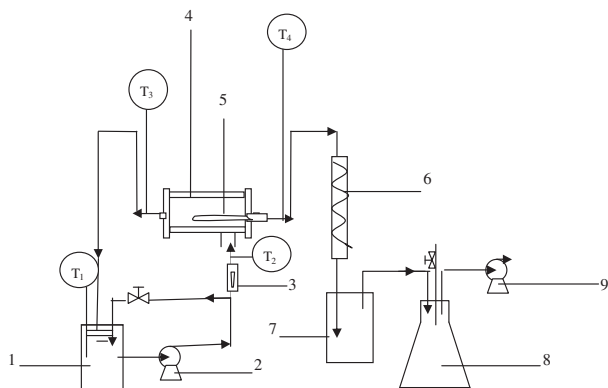


Fig. 2. The experimental setup of vacuum distillation (1. tank for feed liquid, 2. magnetic pump, 3. flowmeter, 4. membrane cisterna, 5. membrane module, 6. drain sleeve, 7. surge flask, 8. collector, and 9. vacuum pump).

In Fig. 2, we can see that there are four temperatures that need to be tested. T_1 represents the temperature of the NaCl solution in a beaker, T_2 is the temperature of the feed NaCl solution, T_3 is reflow water temperature, T_4 is the temperature of permeated water. In this experiment, we recorded the four temperatures in order to analyze them exactly. We used the outside/in permeation model, which is described by Wirth and Cabassud [11].

2.4. The calculation of membrane flux, retention rate, and Reynolds number

The flux is defined as the volume of liquid per unit time and per unit area. We used Eq. (1) to calculate it.

$$J = \frac{V}{ST} \quad (1)$$

where J is flux ($\text{L}/\text{m}^2\text{h}$); V —volume at certain time (L); S —surface area (m^2); T —time (h).

The retention rate was tested by a conductivity meter, and calculated using Eq. (2).

$$R_j = \frac{\rho h - \rho c}{\rho h} \times 100\% \quad (2)$$

where ρh is electrical conductivity of feed-in solution, ρc —electrical conductivity of distillation production.

In this test, we used Eq. (3) to calculate the Reynolds number which is represented by Xu [12]

$$\text{Re} = 5.95DUT^{0.88} \quad (3)$$

where D is diameter of the pipe (cm), U is the flow rate of the liquid (cm/s), T is temperature of the liquid ($^\circ\text{C}$).

3. Results and discussion

3.1. The impact of feed temperature on flux and salt rejection rate

We listed experimental parameters and data for 20 g/L in Table 2, and other concentrations (40 and 80 g/L) of NaCl solution are the same. Figs. 3 and 4 reflect different concentrations and different temperatures. Figs. 3 and 4 show the impact of feed temperature on water flux and rejection rate. We can see that the higher the feed temperature, the greater the water flux and smaller the rejection rate. Because the vapor pressure of seawater increases with temperature, the driving force for mass transfer and thus the flux increased significantly. At a vacuum pressure of 58 cm Hg, the flow rate 5 L/h, and feed temperature 80°C , the water flux through the vacuum MD process could reach $7.8\text{L}/\text{m}^2\text{h}$. The maximum salt rejection rate could reach 99.9%. Contrary to the rejection rate, the water flux decreased with the temperature increasing,

Table 2
Experiment parameters and data for 20 g/L NaCl solution

T_1 ($^\circ\text{C}$)	T_2 ($^\circ\text{C}$)	T_3 ($^\circ\text{C}$)	T_4 ($^\circ\text{C}$)	Vacuum pressure (cm Hg)	Q (L/min)	t (min)	J ($t_1 - t_2$) (ml)	ρh (ms/cm)	ρc ($\mu\text{s}/\text{cm}$)
45	40	28	40	58	5	240	0.1	26.5	19.1
54	50	30	50	58	5	30	0.5	26.4	19.2
65	60	50	60	58	5	30	8.6	26.6	25.5
75	70	50	70	58	5	30	10.2	26.5	51.1
85	80	52	80	58	5	30	26.3	26.5	88.6

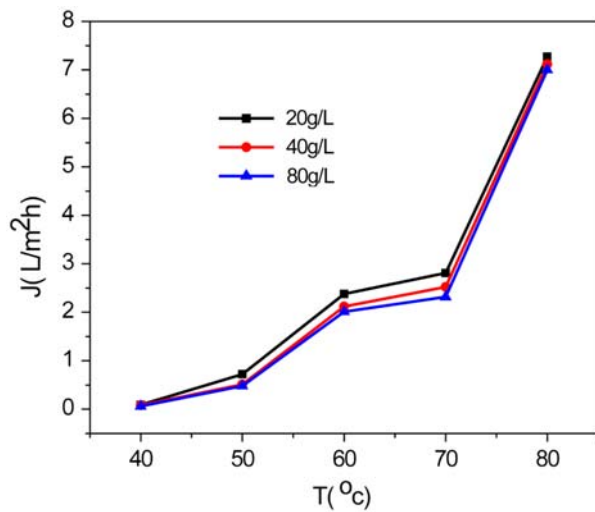


Fig. 3. The impact of feed temperature on water flux.

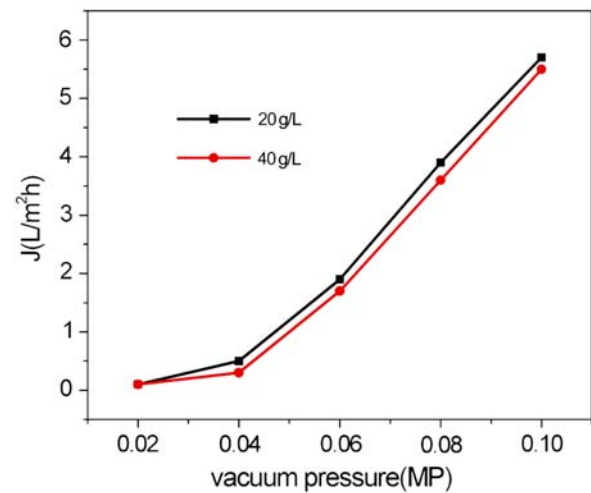


Fig. 5. The impact of vacuum pressure on water flux.

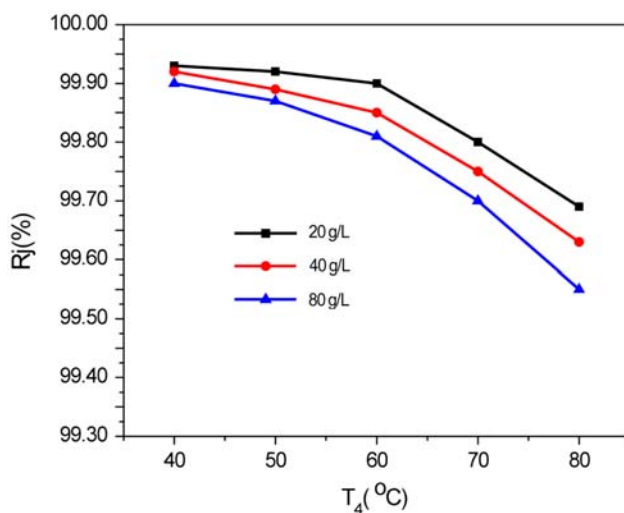


Fig. 4. The impact of feed temperature on Salt rejection rate.

as shown in Fig. 3. The water flux changes unobviously with the initial concentration, and R_j decreases with the initial concentration increase.

3.2. The impact of vacuum pressure on water flux and rejection

The four temperatures ($T_1 - T_4$) are 72, 70, 37, and 70°C, respectively. Simultaneously the pressure of the vacuum side was adjusted and the flow rate was 5 L/min. Water flux and rejection rate were tested in this case. The experimental results are shown in Fig. 5. We can see that with the vacuum side pressure increasing, the water flux significantly increased; however, the

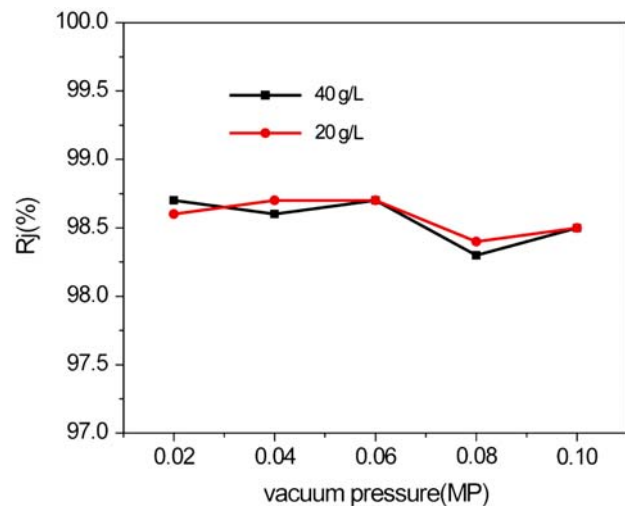


Fig. 6. The impact of vacuum pressure on R_j .

salt retention rate changed a little in Fig. 6. In the low vacuum, the membrane flux increased gradually with increasing the degree of the vacuum pressure. When the vacuum was greater than 0.06 MP, the permeate flux increased rapidly with the increase in the vacuum. In a lower degree, vacuum the water vapor through the membrane was driven from the membrane on both sides. The dynamic vapor pressure through the membrane increased with the increase in the cold side of the vacuum, making changes in the membrane flux as well.

3.3. The impact of flow rate on water flux and rejection

Reynolds number can be used to characterize the fluid flow state, which is represented by $Re = \rho v d / \eta$, where v , ρ , and η , respectively stand for the velocity,

Table 3
Relevant parameters and data for 80°C

T_4 (°C)	Q (L/min)	Flow rate (cm/s)	Re	J (L/m ² h)	R_j
80	1	1.06	2,651	7.05	99.80
80	2	2.12	5,302	7.67	99.85
80	3	3.18	7,953	7.75	99.91
80	4	4.24	10,604	7.76	99.93
80	5	5.3	13,255	7.79	99.95

density, and viscosity of the fluid; d is the characteristic length. Like fluid flow through a circular pipe, d is the diameter of the pipe. The use of the Reynolds number can distinguish whether the fluid flow is laminar or turbulent. Flow liquid is in laminar flow conditions when $Re < 2,000$ and in turbulent state when $Re > 4,000$. We adjusted flow capacity (1–5 L/min). Because the velocity of liquid is difficult to find, we used Eq. (3) to calculate it. The relevant parameters and data are listed in Table 3.

We can see from Table 3 that the water flux J increased with the increase in flow rate. When $Re = 2,651$, the water flux is small because of the water is in laminar and the heat and mass transfer is slow. When the $Re > 4,000$, the liquid flux in turbulent state possess a higher J . This can be explained by the fact that with the increase in flow rate, the heat and the mass transfer of the feed side bounding layer becomes thinning, thus reducing the heat and mass transfer resistance.

4. Conclusions

- (1) The pp hollow fiber membranes made by melt-spinning and stretching method were used for vacuum MD. The micropore size was 0.05–0.3 μm . In this work, the water flux reached 7.8 L/m²h, and the rejection ratio of NaCl reached 99.9%.
- (2) The effects of temperature, concentration, and flow rate of NaCl feed solution on water flux and salt rejection rate were studied. The results indicate that water flux and salt rejection rate

changed a little with the increase in the initial concentration of NaCl solution. Water flux increased apparently with feed temperature and vacuum pressure, and increased slowly with solution flow rate increasing.

Acknowledgments

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