Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Vapor Permeate Characteristics of Membrane Distillation

H. Kurokawa[a]; O. Kuroda[b]; S. Takahashi[b]; K. Ebara[c]
[a] Energy Research Laboratory Hitachi Ltd., Hitachi-shi, Japan [b] Hitachi Research Laboratory Hitachi Ltd., Hitachi-shi, Japan [c] Industrial Process Group Hitachi Ltd., Tokyo, Japan

To cite this Article Kurokawa, H. , Kuroda, O. , Takahashi, S. and Ebara, K.(1990) 'Vapor Permeate Characteristics of Membrane Distillation', Separation Science and Technology, 25: 13, 1349 — 1359
To link to this Article: DOI: 10.1080/01496399008050396
URL: http://dx.doi.org/10.1080/01496399008050396

SEPARATION SCIENCE AND TECHNOLOGY, 25(13-15), pp. 1349-1359 (1990)

VAPOR PERMEATE CHARACTERISTICS OF MEMBRANE DISTILLATION

H. Kurokawa
Energy Research Laboratory
Hitachi Ltd.
1168 Moriyama-cho, Hitachi-shi
316 Japan

K. Ebara
Industrial Process Group
Hitachi Ltd.
6 Kanda-surugadai 4 chome
Chiyoda-ku, Tokyo 101 Japan

O. Kuroda and S. Takahashi
Hitachi Research Laboratory
Hitachi Ltd.
4026 Kuji-cho, Hitachi-shi
319-12 Japan

ABSTRACT

To study the permeate characteristics of membrane distillation considering heat and mass transfers and concentration polarization, theoretical and experimental studies were performed. To understand the effect of concentration polarization, concentrated solutions of LiBr and H_2SO_4 were used as feed. The permeate flux was found to be proportional to the vapor pressure difference between the feed and cooled surface and decreased with increasing concentration because of the concentration and temperature polarization. Furthermore, when considering the heat and mass transfers and concentration polarization, the permeate flux could be estimated accurately.

INTRODUCTION

Membrane distillation uses a membrane separation for liquids accompanied by a phase change. A hydrophobic membrane made from a porous polytetrafluoroethylene (PTFE), polyethylene (PE), or polypropylene (PP) is used because of the sensitivity of these materials to permeation of vapor, not liquid. Since effective permeate rates can be obtained at 50-100°C, membrane distillation is effective for separation and concentration of the solution with nonvolatile materials. Very pure permeate is obtained because of the mist separation effect of the membrane. When PTFE membrane is used for concentration of acids and bases, the membrane materials must have good heat and chemical resistance. Membrane distillation has been studied for desalination systems (1,2), water separation from blood (3), and water purification systems (4).

In previous reports (5,6), it was confirmed that the permeate characteristics of water vapor are affected by the diffusion gap between the membrane and the cooled surface. The permeate flux is proportional to the vapor pressure difference between the feed and cooling surface and is influenced by the porosity and thickness of the membrane and the diffusion gap. It was also confirmed that when the diffusion gap is below 5 mm the heat and mass transfer rates in the condensing channel are controlled by diffusion, and when over 5 mm they are controlled by free convection.

This paper describes the results of fundamental experiments on the permeate characteristics of membrane distillation. Permeate experiments were performed using concentrated LiBr and H_2SO_4 solutions, and the effect of concentration polarization in the feed channel was studied.

THEORETICAL STUDY

Principle of Membrane Distillation

Figure 1 illustrates the principle of membrane distillation. Liquid on the high temperature side of the membrane is vaporized and permeates through the membrane pores. Permeate is obtained by condensation on a cooled plate placed near the membrane. In this method a porous hydrophobic membrane is used because it allows permeation of vapor only, not liquid. The distance between the membrane and the cooling surface is referred to as the diffusion gap.

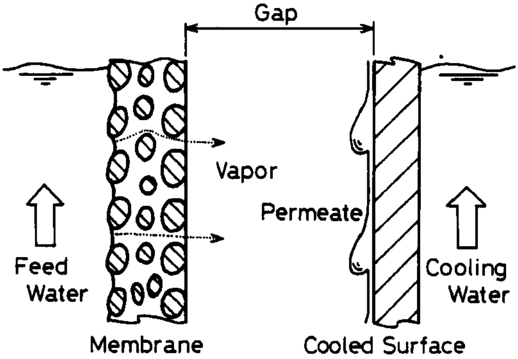

Fig. 1. Separation model of membrane distillation.

Heat and Mass Transfer in the Condensed Channel

In membrane distillation, the vapor from the feed moves through the air and condenses at the cooled surface. The transfer of water vapor is affected by diffusion in the air and free convection due to the concentration difference of vapor. The heat transfer is also affected by heat conduction and free convection. Figure 2 shows the model for heat and mass transfer of membrane distillation.

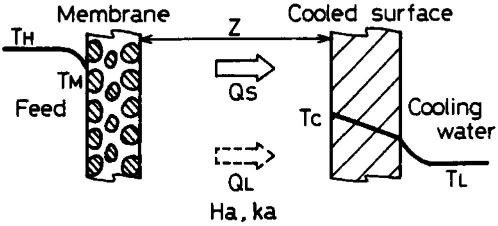

Fig. 2. Mode of heat and mass transfer of membrane distillation.

Heat conduction and diffusion. The heat transfer rate in membrane distillation must supply energy for both latent heat and sensible heat.

$$Q_T = Q_L + Q_S \tag{1}$$

where Q_T is the total heat transfer, Q_L is the rate of change in latent heat, and Q_S is the rate of change in sensible heat. When the diffusion gap is below 5 mm, the heat transfer is controlled by heat conduction. Therefore, the heat transfer rate is expressed using

$$Q_S = ka \cdot \Delta T_{MC}/Z + F \cdot Cp \cdot \Delta T_{MC} \tag{2}$$

$$Q_L = \lambda \cdot F \tag{3}$$

where ka is thermal conductivity; ΔT_{MC} is temperature difference between the surface of the membrane and the cooled surface ($\Delta T_{MC} = T_M - T_C$ where T_M is the temperature of feed on the membrane surface and T_C is the temperature of the cooled surface); Z is the diffusion gap; F is the permeate flux; Cp is the specific heat; and λ is the latent heat.

One-directional diffusion can be applied, and air is stationary. Therefore, the permeate flux, F, is expressed as follows (5):

$$F = \frac{\pi \cdot m}{R \cdot T \cdot Pa} \cdot \left(\frac{1}{\delta/D_m + Z/D} \right) \cdot \Delta P \tag{4}$$

where π is total pressure, m is the molecular weight of water, R is the gas constant, T is temperature, Pa is partial pressure of water, δ is the membrane thickness, D_m is the diffusion coefficient in the membrane, D is the diffusion coefficient in the air gap, and ΔP is the water vapor pressure difference between the feed on the membrane and the cooled surface. In Eq. (4), D_m is expressed as follows:

$$D_m = \epsilon \cdot D/\tau = D \cdot \epsilon^{3.6} \qquad (5)$$

where ϵ is the porosity of the membrane and τ is the tortuosity of the membrane. In Eq. (5), $\tau = \epsilon^{-2.6}$ is estimated from direct contact type membrane distillation experiments. From Eqs. (4) and (5), the permeate flux is expressed as Eq. (6).

$$F = \frac{\pi \cdot m}{R \cdot T \cdot Pa} \cdot \left(\frac{D}{\delta/\epsilon^{3.6} + Z} \right) \cdot \Delta P. \qquad (6)$$

Using Eqs. (1)-(6), T_M, T_C, and F can be calculated when the diffusion gap is below 5 mm.

Free convection. When the gap is over 5 mm, F and Q_L are controlled by free convection. Heat transfer is adjusted by using the Nu (Nusselt) number, Gr (Grashof) number, and Pr (Prandtl) number. In particular, the free convection heat transfer at a gap between two vertical parallel plates is given by the following equation (7):

$$Nu = \frac{c}{(\ell/Z)^{1/9}} \cdot (Pr \cdot Gr)^n \qquad (7)$$

$2.1 \times 10^5 < Gr < 1.1 \times 10^7$ $\quad c = 0.07$ $\quad n = 1/3$
$2.0 \times 10^4 < Gr < 2.1 \times 10^5$ $\quad c = 0.20$ $\quad n = 1/4$

where ℓ is the length of the plates and c and n are constants.

Previously, it was reported (5) that the mass transfer rate by free convection is expressed by an equation similar to Eq. (7) because of the analogy between the heat and mass transfers as follows:

$$Sh = \frac{c}{(\ell/Z)^{1/9}} \cdot (Sc \cdot Gr)^n \qquad (8)$$

$2.1 \times 10^5 < Gr < 1.1 \times 10^7$ $\quad c = 0.07$ $\quad n = 1/3$
$2.0 \times 10^4 < Gr < 2.1 \times 10^5$ $\quad c = 0.20$ $\quad n = 1/4$

From Eqs. (7) and (8), the heat transfer coefficient, Ha, and mass transfer coefficient, km, are calculated. F and Q are calculated by Eqs. (9)-(12).

$$Q_T = Q_S + Q_L \qquad (9)$$

$$Q_S = Ha \cdot \Delta T_{MC}/Z + F \cdot Cp \cdot \Delta T_{MC} \qquad (10)$$

$$Q_L = \lambda \cdot F \tag{11}$$

$$F = km \cdot (C_M - C_C) \tag{12}$$

where C_M and C_C are concentrations of water vapor at the membrane surface and the the cooled surface, respectively. Using Eqs. (7) and (8), T_M, T_C, and F can be calculated.

EXPERIMENTAL STUDY

Experimental Apparatus and Method

Figure 3 shows a flow chart of the experimental apparatus for the membrane distillation. The cell for the experiment had three channels divided by the membrane and a stainless steel plate. The cell size was 240 x 150 mm and the effective membrane area was 1.42 x $10^{-2}m^2$. The membrane used was made of PTFE; its pore diameter was 0.2 μm; and its porosity was about 76%.

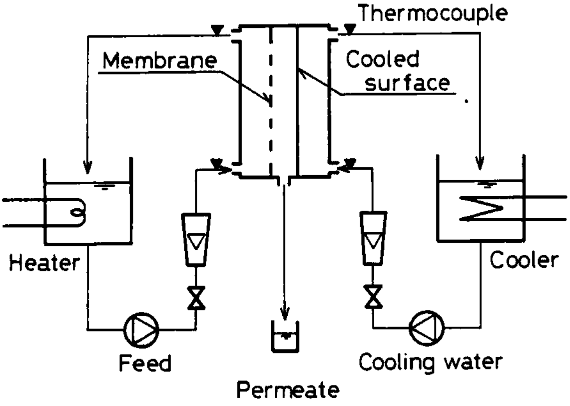

Fig. 3. Schematic diagram of the experimental apparatus.

The high temperature liquid and the water for cooling the plate surface were fed to the respective outer channels, while the condensed water was obtained from the middle channel. The condensed permeate was collected with a saucer. In the experiments, after the temperature and flow rate were stable, the temperatures of the inlet and outlet of the feed and cooling water and the permeate rate were measured. The permeate rate was obtained from the weight of the permeate. As the objective of this report was to study the effect of concentration polarization, parameters such as concentration, temperature, and flow rate of feed were varied.

Experimental Results and Considerations

Figures 4 and 5 show the relationships between the permeate flux, F, and the vapor pressure difference, ΔP, when the concentrations of LiBr solution and H_2SO_4 solution were varied respectively. Since F was proportional to ΔP, Eq. (13) was obtained.

$$F = K \cdot \Delta P \tag{13}$$

where K was a proportionality constant which expressed permeate rate. The F values decreased with increasing concentration of solutions. Furthermore, the purity of permeate was shown as their electric conductivity, σ. The σ of the permeate was below 10 μS/cm. Then, rejection of this method was more than 99.9% considering the concentration of feed solution.

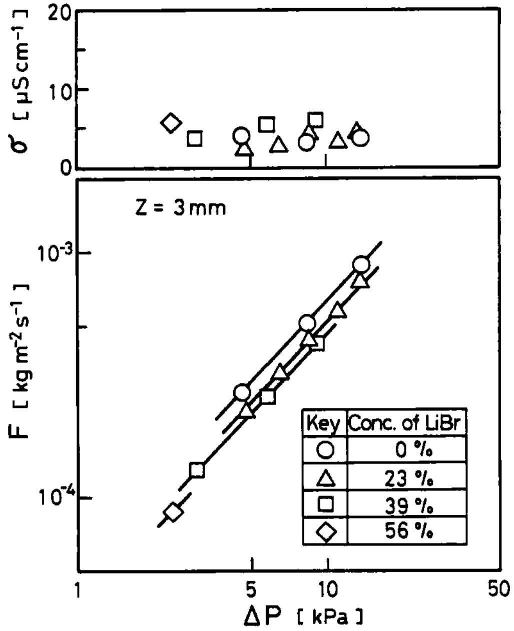

Fig. 4. The relationships between permeate flux, F, and vapor pressure difference between feed and cooled surface when LiBr solution was used as feed.

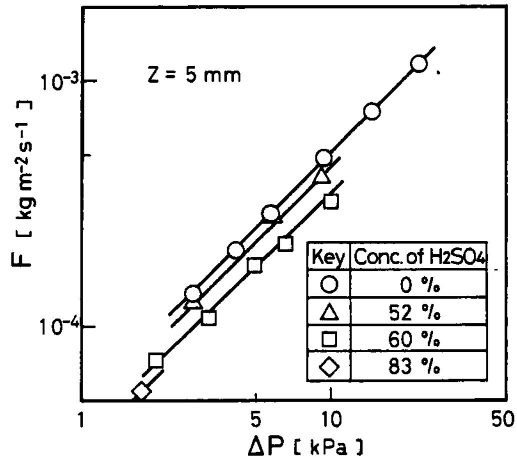

Fig. 5. The relationships between permeate flux, F, and vapor pressure difference between feed and cooled surface when H_2SO_4 solution was used as feed.

Figure 6 shows the relationships between the permeability, K, and concentration of solution, C, when the vapor pressure difference was 10 kPa. K decreased with increasing C. This was due to the temperature and concentration polarization of feed solution, because in these figures ΔP was the vapor pressure difference between the feed and cooling water. Therefore, if temperature and concentration polarization occurred, the real vapor pressure difference would be small versus the ΔP. From these results, it was confirmed that the effects of temperature and concentration polarization increased with increasing concentration of feed. The reason for these phenomena was considered to be the exponential increase in the vapor pressure of the LiBr solution with increasing temperature and concentration.

Figure 7 shows the relationship between the Reynolds number (Re) and the permeability, K, when the concentration of the LiBr solution was varied. The permeate flux slowly increased with increasing Re and the boundary layer was thinned for larger Re. The solid lines in Fig. 7 were calculated using Eqs. (1)-(12). The calculated temperature polarization was obtained using these equations and did not correlate well with the experimental data. This was attributed to the difference in concentration polarization at the upstream surface of the membrane.

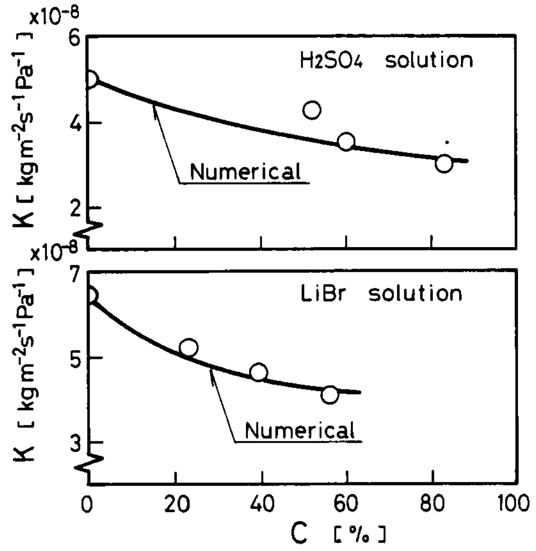

Fig. 6. The relationships between permeability, K, and concentration of feed.

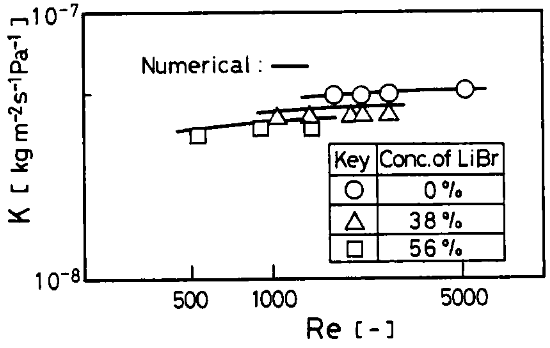

Fig. 7. The relationship between the permeability, K, and Reynolds number, Re, of the feed.

In the membrane separation process, concentration polarization is a major problem. The boundary layer of concentration in the membrane process is shown in Fig. 8. Solute and solvent fluxes, J_S and J_V, are expressed as follows:

$$J_S = X_A(J_S + J_V) - D_{AB} \cdot c \cdot \frac{dX_A}{dx} \tag{14}$$

$$x = 0 \qquad X_A = X_{A1} \tag{15}$$

$$x = lm \qquad X_A = X_{A2} \tag{16}$$

where X_A is the mole fraction of solute, D_{AB} is the diffusion coefficient, c is the mole concentration, and x is the thickness of the boundary layer. Using the conditions of Eqs. (15) and (16), Eq. (14) is integrated as follows:

$$J_S + J_V = c \cdot k \cdot \ln\left(\frac{X_{A2} - X_{A3}}{X_{A1} - X_{A3}}\right) \tag{17}$$

where k is the mass transfer coefficient (k = D/lm), X_{A1} is the mole fraction of solute in feed, X_{A2} is the mole fraction at the membrane surface, and X_{A3} is the mole fraction of the permeate. In the case of membrane distillation, as the rejection is nearly 100%, values of J_S and X_{A3} are zero. Then, the solvent flux, J_V, is expressed as Eq. (18).

$$J_V = c \cdot k \cdot \ln\left(\frac{X_{A2}}{X_{A1}}\right) \tag{18}$$

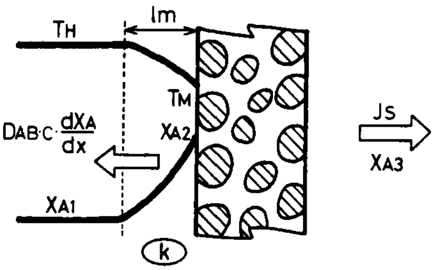

Fig. 8. Model of concentration boundary layer in the membrane distillation.

The effect of concentration polarization has been studied by many researchers for reverse osmosis and ultrafiltration membrane processes. In the turbulent flow region, Dittus-Boelter's equation [Eq. (19)] gives the good fit for the phenomenon (7).

$$Sh = 0.023 \cdot Re^{0.8} \cdot Sc^{1/3} \tag{19}$$

From Eq. (19), km can be calculated and then the concentration of solute at the membrane surface can be calculated by Eq. (18).

Figure 9 shows the relationship between the permeate flux from the experiments and calculations using Eqs. (1)-(19). Regarding all regions of the experiments, a good correlation was obtained between the experimental and numerical results. Then, the permeate flux could be estimated by considering the heat and mass transfers and concentration polarization.

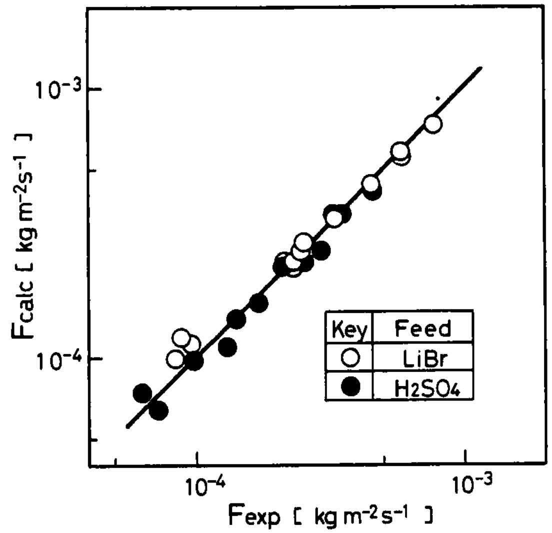

Fig. 9. The relationship between the permeate flux of experiments F_{exp} and of calculations F_{calc}.

CONCLUSIONS

To study the permeate characteristics of membrane distillation considering the heat and mass transfers and concentration polarization, theoretical and experimental studies were performed. The following points were seen:

1. Using concentrated solutions as feed, the permeate flux decreased with increasing concentration because of the concentration and temperature polarization.

2. Accounting for both the heat and mass transfers and concentration polarization, it was confirmed that permeate flux could be estimated accurately for all experimental conditions.

3. Using concentrated solutions of LiBr, electric conductivity of permeate was below 10 μS/cm. Therefore, rejection of this method was more than 99.9%.

REFERENCES

1. Aptel, P., N. Challard, J. Cuny, and J. Neel, J. Memb. Sci. 1, 271 (1971).

2. Gore, D. W., Tech. Proc. W.S.I.A., July 25-29, Honolulu, Hawaii, Vol. III, 1 (1982).

3. Hosoya, N., K. Yokota, T. Koyano, K. Sakai, and M. Tamura, Preprint of the 54th Annual Meeting of the Soc. of Chem. Engrs., Japan, p. 375, Kobe (1989).

4. Ebara, K., H. Kurokawa, A. Yamada, Y. Koseki, and A. Ashida, Proc. of the 15th I.S.T.S., p. 1355, Tokyo (1986).

5. Kurokawa, H., Y. Koseki, A. Yamada, K. Ebara, and S. Takahashi, Kagaku Kogaku Ronbunsyu 14, (3), 330 (1988).

6. Kurokawa, H., Y. Koseki, A. Yamada, K. Ebara, and S. Takahashi, Kagaku Kogaku Ronbunsyu, to be submitted.

7. Kagaku Kogaku Kyokai, "Kagaku Kogaku Binran," 3rd ed., p. 245, Maruzen, Tokyo (1969).