

Pervaporation Study of Water and *tert*-Butanol Mixtures

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ABSTRACT: The permeation behavior of water/*tert*-butanol mixture through Sulzer Pervap2510 hydrophilic poly(vinyl alcohol) membranes was investigated and the effects of feed composition and temperature on separation efficiency of the membranes were studied. The pervaporation experiments were carried out with feed water content varying from 0 to 20 wt % according to the existing industrial needs and with the feed temperature from 60 to 100°C. Over this range, both water flux and separation factor increased with increasing water content and feed temperature. These phenomena may be attributed to (1) the strong interaction between water and the membrane, (2) the decoupling effect of the permeants and the membrane at elevated temperatures,

and (3) the steric hindrance effect of branch chain alcohol. The permeability ratio (the ideal separation factor) of water to *tert*-butanol across the membrane was calculated and found to follow the same relationship with increasing temperature and water content. Both flux and separation factor obtained from the Pervap2510 membrane in this study were much higher than previous reported values, possible causes for which were analyzed. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 4082–4090, 2004

Key words: pervaporation; water/*tert*-butanol mixture; dehydration; membranes; chain

INTRODUCTION

Separation and purification of organic solvents can achieve great economical and environmental benefits in the chemical and pharmaceutical industries. However, many organic solvents are heat sensitive and known to form azeotropes easily, which makes the separation process difficult and challenging.^{1,2} The pervaporation separation process is a potential alternative for the separation and purification of organic solvent mixtures and intermediate pharmaceuticals products because of its unique features of low energy consumption and minimum contamination.

Two transport theories have been proposed for the separation principle of pervaporation: (1) the solution–diffusion mechanism^{3–5} and (2) the pore flow mechanism.^{6–8} The former hypothesizes that the selectivity and permeation rate are governed by the solubility and diffusivity of the feed components permeating across the membrane. The solubility of a feed component in the membrane is determined primarily by the chemical nature of the membrane material and the permeating molecules, and may be qualitatively estimated using the solubility parameter,⁹ whereas the

diffusivity is dependent on chemical and physical factors such as the size and shape of penetrant molecules as well as their mutual interactions with the polymer.¹⁰ The latter theory proposes the transportation across the membrane consisting of three consecutive steps: (1) liquid transport from the inlet of membrane pores to the liquid–vapor phase boundary; (2) evaporation at the phase boundary; and (3) vapor transport from the phase boundary to the pore outlet.^{6,7} Both theories agree that the complicated chemical and physical interactions among feed components as well as among feed molecules and membranes play important roles to determine the overall separation performance.

Performance of pervaporation is dependent not only on the membranes but also on the operating parameters such as feed composition, temperature, and other factors.^{11–14} Because the sorption and diffusion of each component are strongly concentration dependent, changing of feed composition will affect the flux and selectivity in pervaporation. Yeom et al.¹² investigated a homologous series of aqueous alcohol solutions through a poly(vinyl alcohol) (PVA) membrane with various feed compositions and found that the feed composition would directly affect the affinities between feed components and membrane as well as the mutual interactions among the feed components. On the other hand, changing the operating temperature may cause the change of membrane structure and mutual interaction between components, which

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consequently contribute to the change of the mass transport coefficient of components.¹³ Lai et al.¹⁴ provided a detailed study on the effect of feed temperature in the separation of ethanol from water by using a polysiloxaneimide membrane and concluded that the desired separation factor can be achieved by careful control of feed temperature.

In this report, the permeation behavior of water/*tert*-butanol mixtures through a commercial hydrophilic PVA membrane was investigated. *tert*-Butanol was chosen because it is a widely used organic solvent in chemical and pharmaceutical syntheses. Even though the recoveries of other butanols, such as *n*-butanol,¹⁵ 2-butanol from model solutions,¹⁶ or from ABE (acetone-butanol-ethanol) fermentation broths^{17,18} by pervaporation have often been reported, only limited amounts of data are available on the *tert*-butanol recovery. Except for the recent work of Gallego-Lizon et al.,¹⁹ most existing reports^{20,21} on *tert*-butanol were based on the membranes fabricated or modified individually in the laboratory, which may probably be ascribed to commercial pervaporation membranes being seldom available a decade ago. Therefore, the purpose of this study was to investigate *tert*-butanol dehydration by using a commercial polymeric membrane and to examine the dehydration kinetics and separation performance as a function of membrane materials, feed composition, and operating temperature.

BASIC THEORY

The separation characteristics of pervaporation are far more complex than liquid and gas separation because the former includes two phases: vapor and liquid. It involves (1) the physicochemical properties of feed mixtures and their own interactions, (2) the affinity of permeants toward the macromolecules that constitute the membrane, and (3) the physical structure of the membrane. Thus, the solubility and diffusivity are not constants but are strongly dependent on feed concentration and temperature. According to the solution-diffusion mechanism, the membrane permeability coefficient is a product of the solubility coefficient and diffusivity coefficient.

The experiment system, as shown in Figure 1, is a closed pervaporation system. The feed side is at a liquid-vapor equilibrium status for a given temperature and the pressure as a driving force on the feed side is equal to the saturated vapor pressure. The basic transport equation for the component *i* can be expressed as

$$J_i = \left(\frac{DS}{\delta} \right)_i (P_s X_{iS} - P_p Y_i) \quad (1)$$

where *J* is the permeate flux; δ is the membrane thickness; *P* represents the total pressure; *D* and *S* are the

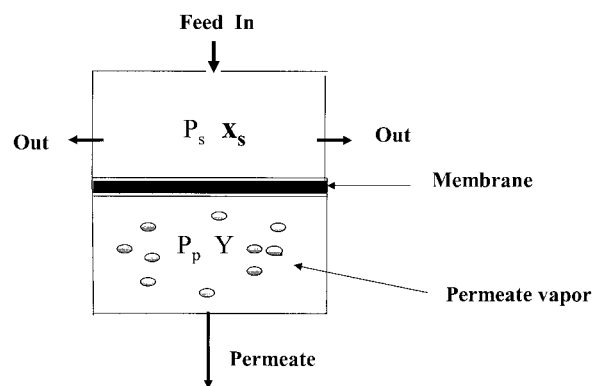


Figure 1 Transport in the pervaporation process.

diffusivity and solubility coefficients, respectively; *X* and *Y* are the mole fractions of a specific component in the feed and permeate sides, respectively; the subscript *i* is the corresponding component and the subscripts *s* and *p* denote the saturated vapor (in equilibrium with the feed solution), respectively; (DS/δ) is known as the membrane permeance²²; and $(P_s X_{iS} - P_p Y_i)$ is the driving force expressed in terms of partial pressure difference for the component *i*.

In a binary system, eq. (1) can be rewritten as follows:

$$J_1 = \left(\frac{DS}{\delta} \right)_1 (P_s X_{1S} - P_p Y_1) \quad (2)$$

$$J_2 = \left(\frac{DS}{\delta} \right)_2 (P_s X_{2S} - P_p Y_2) \quad (3)$$

The subscripts 1 and 2 in the discussed system refer to water and *tert*-butanol, respectively. The composition of permeate depends on the fluxes of all species. The mole fraction of water in the permeate may be expressed as

$$Y_1 = \frac{J_1}{J_1 + J_2} \quad (4)$$

whereas the mole fractions in the feed and permeate sides obey the following relationships, respectively:

$$X_{1S} + X_{2S} = 1 \quad (5)$$

$$Y_1 + Y_2 = 1 \quad (6)$$

By rearranging eqs. (2) to (6), *M*, the permeability ratio of water to *tert*-butanol across the membrane, may be expressed as

$$M = \frac{Y_1^2 - Y_1 - Y_1 X_{1S} F + F Y_1}{Y_1^2 - Y_1 - Y_1 X_{1S} F + F X_{1S}} \quad (7)$$

or

$$M = \frac{Y_1 X_{2S} - Y_1 Y_2 / F}{Y_2 X_{1S} - Y_1 Y_2 / F} \quad (8)$$

where

$$M = \left(\frac{DS}{\delta} \right)_1 / \left(\frac{DS}{\delta} \right)_2 \quad (9)$$

$$F = \frac{P_s}{P_p} \quad (10)$$

where F is the ratio of the saturation vapor pressure to permeate vapor pressure. Equation (8) can be further simplified as follows if the vapor pressure in the permeate side is equal to zero and F is approaching infinity according to eq. (10):

$$M = \frac{Y_1 / Y_2}{X_{1S} / X_{2S}} \quad (11)$$

Under this special condition, M is called as the ideal separation factor based on the compositions in the saturated vapor. The saturation vapor composition (in mole fraction) on the feed side can be obtained from the vapor-liquid equilibrium data at the given feed composition and temperature.²³

EXPERIMENTAL

Materials

An analytical grade *tert*-butanol (supplied by Mallinckrodt Chemical) was used together with deionized water to prepare the solution. The *tert*-butanol is a colorless liquid with characteristic odor and is highly flammable. Its boiling point is 82.8°C at atmospheric pressure. It forms an azeotrope with water at 79.9°C with the weight concentration of 88.2%. The binary system studied is a water/*tert*-butanol mixture with water content in the range of 0–20 wt %.

Membrane

A commercially available membrane, Pervap2510 (Sulzer Chemtech GmbH, Neunkirchen, Germany), was used in this study. It is a type of polymeric dehydration membrane for the dehydration of neutral solvents and is applicable for operations at less than 100°C with feed water content less than 20 wt %. Similar to common dehydration membranes, its active layer is composed of special crosslinked PVA with a

thickness of 0.5–2 μm lying on a porous polyacrylic nitrile (PAN) substrate on a polyphenylene sulfide (PPS) supporter. To achieve optimal and reproducible results, the Pervap2510 membrane was conditioned at 95°C for about 2–3 h with a water and *tert*-butanol mixture at water weight content of 5–10% before performance evaluation.

Apparatus for pervaporation experiments

The pervaporation experiments were conducted in a laboratory-scale Sulzer pervaporation unit, supplied by Sulzer Chemtech. Figure 2 shows the schematic layout where the membrane was placed in a stainless-steel cell with an inner diameter of 15.24 cm and an estimated surface area of 178 cm².

The prepared water and *tert*-butanol mixture of about 2 L was fed into the solvent-mixture tank, which has a maximum volume of 2.5 L. The tank was heated by a circulated heating bath with the aid of a single-stage rotary vane pump. The flow rate was set to 85 L/h and the temperature was selected in the range from 60 to 100°C. The feed mixture entered the cell from the top center opening, flowed through the thin horizontal channel, left the cell through the side opening, and flowed back to the tank (similar to Fig. 1). Two thermocouples were placed to monitor and ensure that temperature differences within the cell were less than 1 to 2°C. On the downstream side, a vacuum pressure (P_p) of about 3 mbar was applied and the permeants were condensed by two cold traps filled with the dry ice/acetone mixture in series to ensure all permeants were fully collected.

Flux, separation factor, and m calculation

To measure the fluxes and the separation factor of water to *tert*-butanol, both the feed and permeate samples were collected in a fixed interval (normally 1 h) after the system was allowed to stabilize for 2 h. The flux was determined by weighing the permeate mass using a Mettler Toledo balance and divided it by the product of the interval time and membrane area. An HP 6890 GC (gas chromatograph; Hewlett-Packard, Palo Alto, CA) equipped with a HP-INNOWAX column (crosslinked polyethylene glycol) and a TCD detector is used to analyze the compositions of the feed and the permeate samples. The separation factor α is defined by

$$\alpha = \frac{y_1 / y_2}{x_1 / x_2} \quad (12)$$

where y and x are the weight fractions of components in the permeate and feed, respectively. The feed composition used in separation factor calculation is the

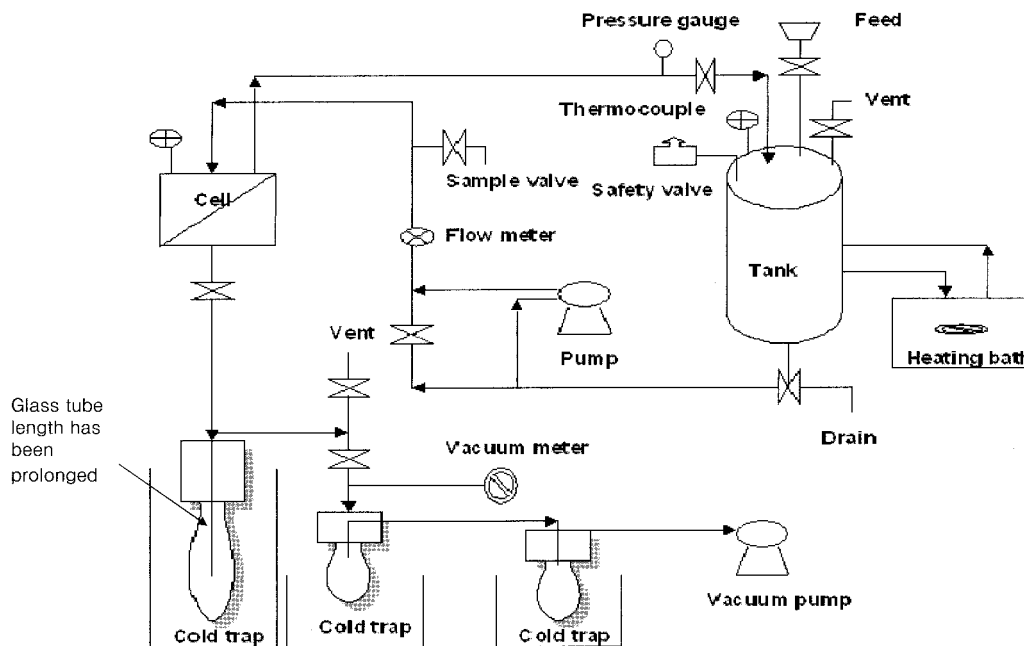


Figure 2 Schematic of the lab-scale pervaporation system.

average value of the feed compositions for the beginning and the end of the time interval of permeate sample collection.

The calculation of M is elucidated as follows. The fugacities f of each component in both phases are equal when the liquid and vapor phases are in equilibrium:

$$f_1^o = P_s X_{1s} = f_1^l = P_1^o \gamma_1 X_1 \quad (13)$$

$$f_2^o = P_s X_{2s} = f_2^l = P_2^o \gamma_2 X_2 \quad (14)$$

where P^o is vapor pressure of the pure component and γ is the activity coefficient. P^o can be estimated with the aid of the Antoine equation²³

$$\log_{10} P^o = A - B/(T + C) \quad (15)$$

where A , B , and C are Antoine constants. Their values vary only slightly with temperature and pressure.²³ The activity coefficient γ can be predicted using the Wilson equation, as follows²³:

$$\ln \gamma_1 = -\ln(X_1 + \Lambda_{12}X_2) + X_2 \left(\frac{\Lambda_{12}}{X_1 + \Lambda_{12}X_2} - \frac{\Lambda_{21}}{\Lambda_{21}X_1 + X_2} \right) \quad (16)$$

$$\ln \gamma_2 = -\ln(X_2 + \Lambda_{21}X_1) - X_1 \left(\frac{\Lambda_{12}}{X_1 + \Lambda_{12}X_2} - \frac{\Lambda_{21}}{\Lambda_{21}X_1 + X_2} \right) \quad (17)$$

where Λ_{12} and Λ_{21} are the Wilson parameters. In addition, the total vapor pressure can be described as

$$P_s = P_1^o \gamma_1 X_1 + P_2^o \gamma_2 X_2 \quad (18)$$

Combined with eqs. (13), (14), and (18), the values of X_{1s} , X_{2s} , and P_s can be obtained. Therefore, F can be calculated from eq. (9) and M can be obtained according to eq. (8).

RESULTS AND DISCUSSION

Effects of feed composition on membrane performance

Figure 3 displays the water concentration in the retentate against time at different feed temperatures and shows that the amount of water in the retentate decreases with time for all temperatures because of its being continuously removed during the experiments. It should be noted that the initial water concentrations shown in Figure 3 are less than 20% because water permeated through the membrane and was removed continuously during the 2-h stabilization period. More water was removed at higher feed temperatures, and thus the initial water concentration decreased.

The flux and separation factors are strongly dependent on the feed composition so that they are plotted as a function of water concentration in the retentate. Figure 4 indicates that the flux increases with an increase in water content in the retentate. This trend is consistent with the literature when using hydrophilic membranes for dehydration of aqueous solutions²⁴⁻²⁶

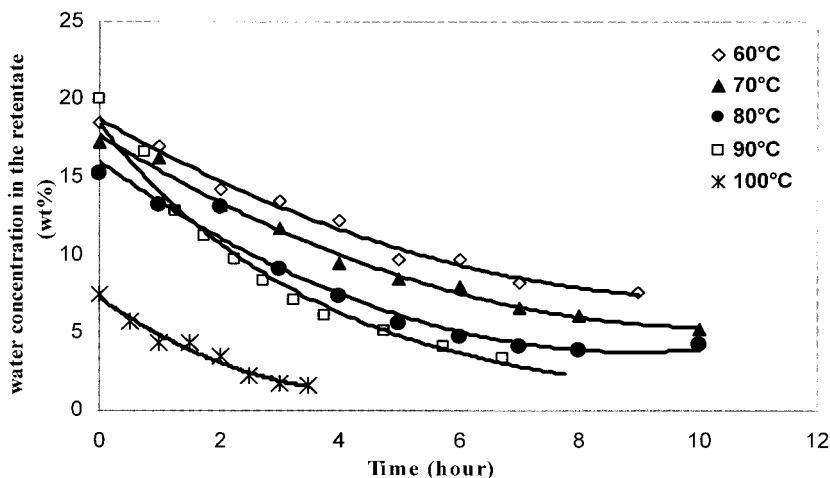


Figure 3 Water concentration in the retentate versus time for the dehydration of a *tert*-butanol/water mixture with PERVAP 2510 membranes at different temperatures.

and this phenomenon can be explained by the interaction between the polymeric membrane and the permeates. PVA is a hydrophilic material and has relatively high polarity groups, resulting in strong interactions with water through hydrogen bonding. As the water content increases, more water molecules can be sorbed into the membrane, thus swelling (plasticizing) the membrane; as a consequence, the permeants are able to pass through the membrane more easily, leading to the flux increase.

It is very interesting to note that the separation factor increases with an increase in water concentration when the temperature is higher than 60°C, as observed in Figure 5. Normally, the permeation rate increases while the separation factor decreases.^{25,26} In the current system, the gross interactions constitute three interactions: (1) membrane and water, (2) membrane and alcohol, and (3) water and alcohol.

When the first two interactions are stronger, more water or alcohol will be sorbed into the membrane because of better affinity among the permeants and the membrane. If the interaction of water and alcohol is overwhelmingly strong, it leads to a coupling effect that will result in the flux increase and separation factor decrease. A simple approach to estimate the interaction between materials is the solubility parameter difference. The solubility parameters δ_{sp} ($\text{cal}/\text{cm}^3)^{1/2}$ of the water, *tert*-butanol, and PVA membrane are 23.4, 10.6, and 19.1, respectively.²⁷ The difference in solubility parameter between each pair is in the order of water/*tert*-butanol > *tert*-butanol/membrane > water/membrane, indicating the water and membrane pair has the strongest interaction because they have the closest solubility parameter compared to that of the other pairs. Thus, high water content not only results in membrane

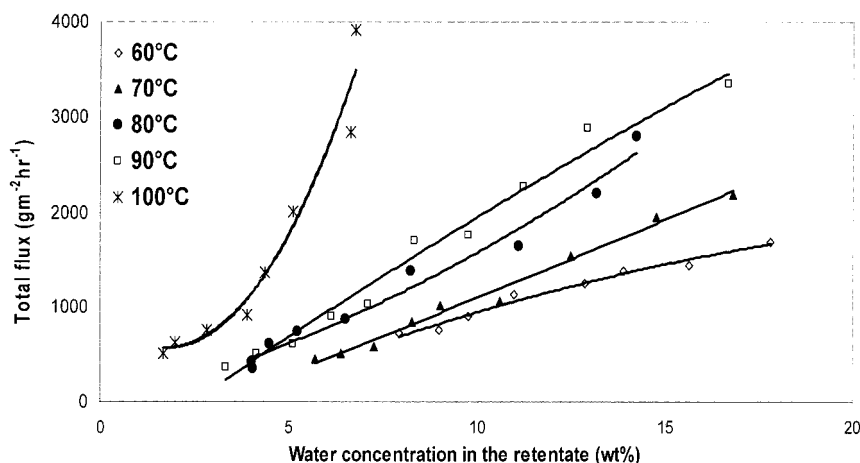


Figure 4 Total flux versus water concentration in the retentate for the dehydration of a *tert*-butanol/water mixture with PERVAP 2510 membranes at different temperatures.

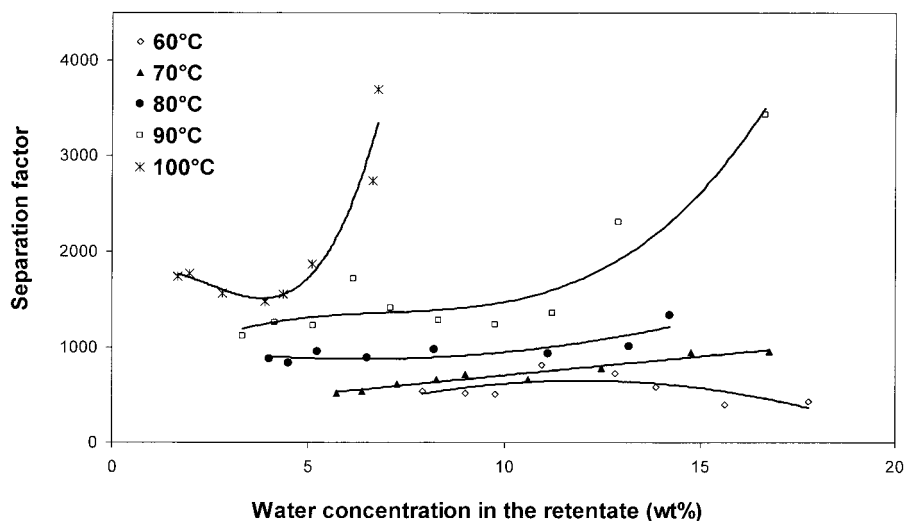


Figure 5 Separation factor versus water concentration in the retentate for the dehydration of a *tert*-butanol/water mixture with PERVAP 2510 membranes at different.

swelling but also creates preferential transportation of water across the membrane.

Effects of feed temperature on membrane performance

Generally, the solubilities, the interactions among the permeants and the membrane, and the membrane free volume vary with temperature. The degree of their individual changes with increasing feed temperatures and their relativity govern the flux and separation factor.

Because a polymer membrane is composed of macromolecules, the random thermal motion of polymeric chains in the amorphous regions produces the free volume. As the temperature increases, the thermal

motion of polymer chains becomes more violent, which brings about an increase in free volume within the membrane. In addition, the increased thermal motion of the permeant molecules at elevated temperatures leads to decoupling of interaction among the permeants. As a result, a high feed temperature may not only increase the overall flux but also enhance the preferential transportation of the permeant that has strong interactions with the membrane.

Figures 6 and 7 show that a higher feed temperature yields a higher separation factor as well as a higher flux. Similar phenomena have been reported for other solvent systems.²⁸⁻³¹ However, the current case cannot be simply explained by the free volume increase at high temperatures. The other reason may be the in-

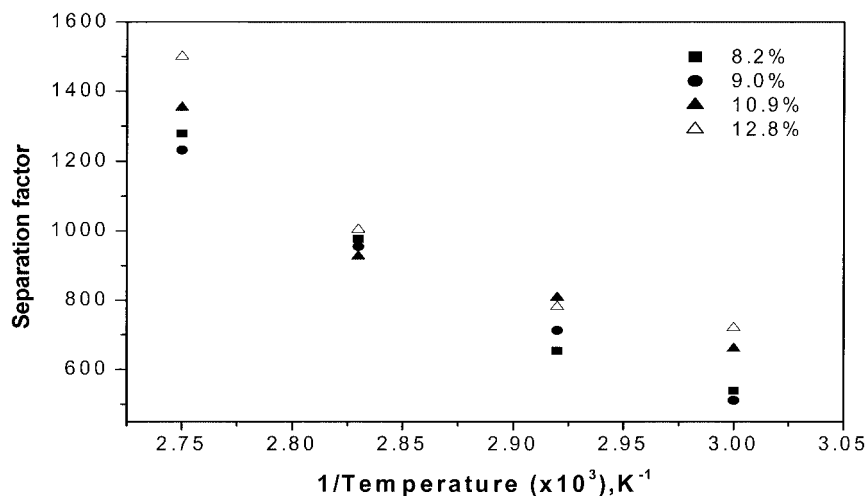


Figure 6 Separation factor versus feed temperature for the dehydration of a *tert*-butanol/water mixture with PERVAP 2510 membranes at different feed compositions.

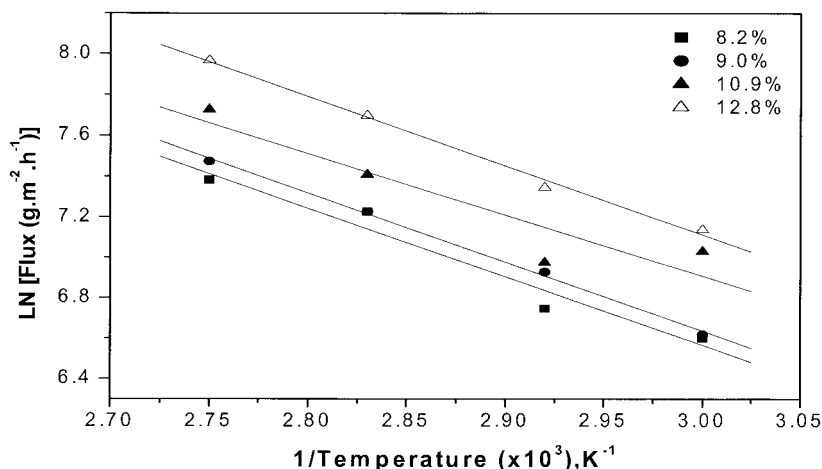


Figure 7 Total flux versus feed temperature for the dehydration of a *tert*-butanol/water mixture with PERVAP 2510 membranes at different feed compositions.

creasing difference in the degree of interaction between water/membrane and *tert*-butanol/membrane pairs at elevated temperatures, which results in the preferential transportation of water. In addition, the steric structure should be taken into account for the diffusion of branch chain alcohol. Although the thermal motion (diffusivity) of permeate molecules accelerates with an increase in temperature and the hydrogen bonding between permeants and membrane molecules become weakened, the diffusion of *tert*-butanol molecules is impeded because of its branch structure and the corresponding steric hindrance effect. This makes its diffusion slower than that of water molecules, resulting in an increase in the separation factor.

An alternative examination on the separation factor, temperature, and water relationship is attempted based on the basic theory discussed earlier. Table I summarizes and compares the values of M calculated from eq. (8) with the experimental values of separation factor α , calculated from eq. (12) at given water concentrations and temperatures. Because M is the permeability ratio of water to *tert*-butanol across the membrane, Table I shows that M and α follow the

TABLE I
Comparison of M Value with Experimental Separation Factor at Different Water Concentrations and Operating Temperatures for the Pervap2510 Membrane

Temperature (°C)	Water concentration (wt %)	M value	Separation factor
60	13.87	1070	618
	9.73	726	509
	7.92	699	539
80	13.18	1079	1001
	9.09	780	876
	8.23	825	977

same trend and their values increase with increasing temperature and water content, suggesting the validity of our previous arguments.

It is important to point out that our findings are different from those reported by Gallego-Lizon et al.¹⁹ where they used the same Pervap2510 membrane but showed lower fluxes than ours and their separation factor decreased with increasing feed temperature. One of the possible reasons for the discrepancy may be attributed to the efficiency of cold traps. In our early experiments, we obtained similar and comparable results as theirs, as shown in Figure 8, when we used the as-purchased cold trap device and used dry ice pellets as the cooling medium. However, a much higher flux was obtained after (1) the glass cooling tube in the cold trap was lengthened from 9 to 19 cm (shown in Fig. 2), (2) the dry ice pellets were replaced by a dry ice and acetone mixture, and (3) the second cold trap was installed. The dry ice and acetone slush can wet the glass tube surface much more effectively and thus significantly enhance vapor condensation. (For reader information, the dry ice and acetone slush has the equilibrium point of -78°C .³²)

The temperature dependency of the permeation rate can be expressed by an Arrhenius equation^{4,5,8}:

$$J = J_0 \exp(-E_p/RT) \quad (19)$$

where J_0 is the preexponential factor, E_p is the apparent activation energy of permeation, and T is the operating temperature. The apparent activation energy for permeation may be calculated from the plot of total flux versus feed temperature at different feed compositions, as shown in Figure 7. The average value of E_p determined from the slopes is 27.4 kJ/mol.

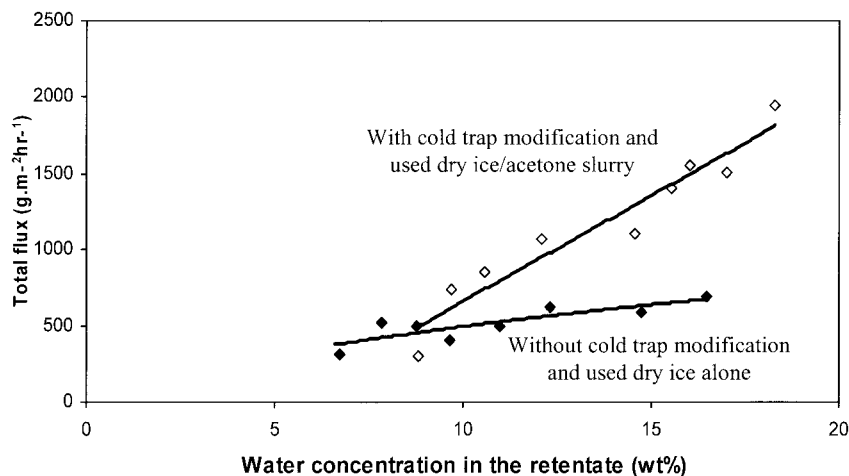


Figure 8 A comparison of cold trap efficiency and cooling medium on total flux ($T = 60^{\circ}\text{C}$, vacuum = 3 mbar, \blacklozenge before cold trap modification and used dry ice, \diamond after cold trap modification and used dry ice plus acetone).

CONCLUSIONS

The dehydration of water from water and *tert*-butanol mixtures was investigated by using Pervap2510, a commercially available membrane. The influence of feed composition and operating temperature on permeation behavior was investigated in a range of feed water concentrations (0–20 wt %) and operating temperatures (60–100°C). For the Pervap2510 membrane, both the flux and separation factor increase with feed water concentration. Hypotheses were offered to explain the experimental results from the standpoint of solubility change with temperature, solubility parameter difference, free volume within the membrane, affinity among permeants and the membrane, and steric hindrance effect of *tert*-butanol molecules. A good agreement was found in a comparison of the ideal and experimental separation factors as a function of temperature and water content. Because the Pervap2510 membrane has rather high water flux and selectivity, it holds promise for future industrial applications.

NOMENCLATURE

- A, B, C Antoine constants
- D diffusivity coefficient
- f fugacity
- f_1^v fugacity of water in vapor phase
- f_2^v fugacity of *tert*-butanol in vapor phase
- f_1^l fugacity of water in liquid phase
- f_2^l fugacity of *tert*-butanol in liquid phase
- F ratio of the saturation vapor pressure to the permeate vapor pressure
- J_i permeate flux of component i
- J_1 permeate flux of water
- J_2 permeate flux of *tert*-butanol

- M permeability ratio of water to *tert*-butanol defined in eqs. (7) and (9)
- P° vapor pressure of pure component defined in eq. (15)
- P_1° water vapor pressure
- P_2° *tert*-butanol vapor pressure
- P_s total pressure of the saturation vapor defined in eq. (18)
- P_p total pressure of the permeate vapor
- S solubility coefficient
- T temperature
- X molar fraction
- X_1 molar fraction of water in the feed liquid
- X_{1s} molar fraction of water in the saturated vapor
- X_2 molar fraction of *tert*-butanol in the feed liquid
- X_{2s} molar fraction of *tert*-butanol in the saturated vapor
- Y_1 molar fraction of water in the permeate
- Y_2 molar fraction of *tert*-butanol in the permeate

Greek letters

- α separation factor
- δ membrane thickness
- δ_{sp} solubility parameter
- γ activity coefficient
- γ_1 activity coefficient of water defined in eq. (16)
- γ_2 activity coefficient of *tert*-butanol defined in eq. (17)
- Λ_{12} Wilson parameter
- Λ_{21} Wilson parameter

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