

Separation of Ethanol from Dilute Aqueous Solution by Pervaporation Process: Influence of Operating Parameters on Flux and Selectivity

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ABSTRACT

Pervaporation can achieve separation of volatile compounds from dilute aqueous solutions due to its high separation efficiency compared to processes such as distillation. Variations of operating parameters such as feed temperature, permeate-side pressure and volumetric feed flow rate can affect permeate flux and selectivity. The separation of ethanol from a 2 %w/w aqueous solution was done using a thin (10 μm) polydimethylsiloxane (PDMS) membrane. Total flux and ethanol selectivity were measured at Reynolds numbers between 500 to 2500, feed temperatures of 30, 40, 50 and 60 °C and permeate-side pressures of 1, 10, 20 and 40 mmHg. Volumetric flow rate was shown to have no significant effect on either total flux or ethanol selectivity whereas feed temperature and permeate-side pressure had highly significant effects.

KEYWORDS

Pervaporation, Flux, Selectivity, Separation, Membrane.

1. INTRODUCTION

Ethanol production from lignocellulosic materials such as forage has attracted interest during recent years as a possible alternative to corn-based ethanol processes [1-3]. The composition of such material typically includes about 33 %w/w cellulose and 28 %w/w hemicelluloses [4], which can be converted to glucose and xylose, respectively, by enzymatic hydrolysis. Although the conversion of cellulosic materials to fermentable sugars is more complex than for starch-rich materials, the potential supply of cellulosic materials is far greater [5]. Glucose and xylose can be fermented to ethanol by certain microorganisms but the fermentation is heterogeneous and the efficiency of ethanol production is low. Belkacemi et al. [6] showed that ethanol concentrations of only 2 %w/w could be produced from forage.

Distillation is the oldest unit operation for separating ethanol from aqueous ethanol-water mixtures. Leeper et al. [7] have shown, however, that distillation is not economical when the initial ethanol concentration in the feed is below 5 %w/w.

Pervaporation, a more recent technique for separating volatile substances from dilute aqueous solutions, is characterized by high separation efficiency at low energy input. An ethanol-water solution can be concentrated

significantly by contacting it with a suitably selective membrane on the outside of which a vacuum is created. By maintaining a vacuum pressure below the vapor pressure of the solute to be concentrated and higher than the vapor pressure of water at that temperature, volatile solutes such as ethanol may permeate through the membrane selectively than water. Greater difference in vapor pressures, higher will be concentration of the more volatile component in the permeate. Since the vapor pressures of ethanol and water are both strongly dependent on temperature [8], permeate-side pressure and feed temperature both significantly affect permeate flux and ethanol selectivity.

Currently, industrial applications of pervaporation are grouped into two: one is dehydration of alcohols and other organic solvents using hydrophilic or charged polymeric membranes and the other is removal of small quantities of volatile organic compounds from water using hydrophobic membranes [9]. For polymeric pervaporation membranes, extensive research was performed to find an optimized membrane material having selective interaction with a specific component of feed mixture to maximize separation performance in items of separation factor, flux and stability. Several types of alcohol permselective membrane materials have been described in literature [10-14]. Silicone containing polymers were generally found to

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exhibit good organophilicity and silicone rubber (mainly polydimethylsiloxane) based membranes were most investigated for separation of organic aqueous mixtures such as alcohols, ketones, phenols and chlorohydrocarbones [9, 13, 15].

In order to scale up the experimental results obtained in the laboratory for industrial applications, certain process parameters (feed temperature, permeate pressure and hydrodynamic conditions) need to be optimized and reliable mathematical models are necessary to predict the influence of the key parameters. The main objective of this work was to study the effects of various operating parameters such as feed temperature, permeate-side pressure and Reynold's number (volumetric flow rate) on the total flux and ethanol selectivity of a PDMS membrane based pervaporation process with 2 %w/w aqueous ethanol.

2. THEORY

Mass transfer through a non-porous membrane by pervaporation has been described as a three-step mechanism by Binning *et al.* [16]. In the first step, ethanol accumulates at the membrane interface where it is sorbed. In the second step, permeate diffuses into the membrane under the pressure gradient between the inside and outside of the membrane. In the third step, permeate desorbs from the membrane as vapor. This model is called the sorption-diffusion or solution-diffusion model.

The first step implies the formation of a concentration profile layer of increased solute concentration on the membrane surface. Concentration profile layer thickness may be determined by initial solute concentration in the feed solution and the volumetric flow rate on the inside of the membrane. Since some substances are absorbable or solvable into a polymer if they have molecular or thermodynamic similarities to that polymer [17-19], organic substances may be expected to be favored over water in the sorption of a solution through a hydrophobic membrane. An important factor controlling the relative solubility of a solute and water in a membrane is the Hildebrand or solubility parameter [17]. When the solubility parameter value of the solute is closer than that of water to the solubility parameter of the membrane, the solute should have a greater sorption affinity for that membrane than water has.

In the second step, permeate moves through the membrane free volume and exits on the outside of the membrane. Permeation rate in pervaporation obeys Fick's equation. The membrane molecular structure may be the most important factor in permeate diffusion into the membrane. A polymeric membrane with hydrophobic groups should be suitable for ethanol separation from an ethanol-water mixture. Ethanol may move through the membrane free volume, which is created by the segmental motion of polymer chains, while water should have more difficulty because of the incompatibility of its hydrogen-

bonding behavior with the hydrophobic groups in the polymer chains. Greater segmental motion of polymer chains means greater free space and hence faster ethanol diffusion through the membrane.

The above theories imply that ethanol should be relatively concentrated on the permeate-side of a membrane having a greater capacity to absorb ethanol than water. An ethanol-selective membrane should have hydrophobic groups in its molecular structure to interfere with water sorption and chain flexibility (or polymer chain segmental motion) high enough to provide space for ethanol diffusion. Such a membrane would be characterized by the ability to create a permeate-side ethanol concentration higher than the feed solution ethanol concentration. Ethanol selectivity can be calculated by the following equation:

$$\alpha_{EtOH} = \frac{(y_i/y_j)}{(x_i/x_j)} \quad (1)$$

where y_i , y_j , x_i , and x_j refer to the mass fractions of ethanol (i) and water (j) on the permeate (y) and feed solution (x) sides, respectively.

In recent years, many ethanol selective polymeric membranes have been developed to separate ethanol from water-ethanol mixtures. Microporous polytetrafluoroethylene (PTFE) and polypropylene (PP) and non-porous silicone composites such as silicon rubber (SR), polydimethylsiloxane (PDMS), polymethoxysiloxane (PMS) and polyoctylmethylsiloxane (POMS) have shown good performance in separating ethanol [20-24]. Among these, polydimethylsiloxane (PDMS) has shown the highest selectivity for ethanol [20]. This polymer has two symmetrical hydrophobic methyl groups in each repeating unit, an amorphous molecular structure and a rubbery state at ambient temperatures ($T_g = -123$ °C) and is thus highly flexible. The solubility coefficient for PDMS is $15.5 \text{ MPa}^{1/2}$ whereas these values for ethanol and water are 26 and $47.9 \text{ MPa}^{1/2}$, respectively [25].

As in all membrane separations, permeants in pervaporation pass through two resistances in series, namely the concentration or polarization layer and the membrane. The concentration layer thickness and membrane resistance may both determine the permeation rate. Varying the feed volumetric flow rate can change concentration layer thickness, increasing flow rate to decrease concentration layer thickness and produce higher flux. Karlsson and Tragardh [26] have studied the effect of Reynolds number on the separation of some aqueous volatile substances, more specifically on their permeation rates and polarization layer thickness using PDMS membrane, observing that for some, but not all volatile substances, the calculated boundary (polarization) layer mean thickness decreased as Reynolds number increased,

resulting in increased permeation rates.

A liquid-to-vapor phase transition front is established inside the membrane, making mass transfer through the membrane a combination of liquid and vapor permeation in series [27, 28]. Mass transfer through the membrane system may be described schematically as shown in Figure 1.

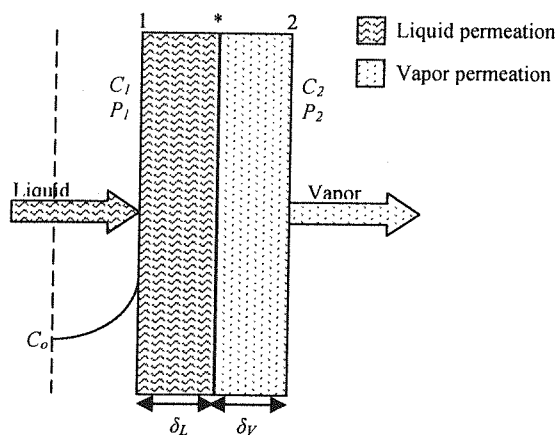


Figure 1: Schematic of mass transfer through the membrane in pervaporation process (P_1 , P_2 , C_1 , and C_2 are pressures and concentrations on the feed and permeate sides of the membrane, P^* , C^* , and C_0 are vapor pressure, concentration at the liquid-vapor interface and initial feed concentration and δ_L and δ_V are liquid and vapor permeation depths in the membrane).

The pressure difference between the feed side and the permeate-side of the membrane creates a pressure gradient within the membrane allowing permeants to enter the membrane as liquid. When the pressure encountered within the membrane is equal to vapor pressure (point “*”) permeants pass to the vapor state immediately and mass transfer proceeds as vapor permeation. Pressure profile and vapor permeation depth (δ_V) vary with changing permeate-side pressure (point 2). Based on the pore flow model, Matsuura and coworkers [29-31] have proposed a new pervaporation mass transfer model for single component dependant on permeate-side, feed side and vapor pressures and on other physical quantities. In this model, permeate quantity (Q) is a combination of mass transfer in the liquid (first term) and vapor (second term) spaces, i.e.,

$$Q = \frac{A}{\delta} (P_1 - P_*) + \frac{B}{\delta} (P_*^2 - P_2^2) \quad (2)$$

where constants A and B include several physical quantities and δ (equal to $\delta_L + \delta_V$) is membrane thickness.

They also calculated the relative liquid transport depth (δ_L) as a function of feed side, permeate side and vapor pressures as follows:

$$\frac{\delta_L}{\delta} = \frac{A(P_1 - P_*)}{A(P_1 - P_*) + B(P_*^2 - P_2^2)} \quad (3)$$

In equation 2, the liquid space pressure profile changes linearly with feed side pressure whereas in the vapor space it decreases as a second order function of permeate side pressure. High feed side and/or low permeate side pressures create a higher gradient force, resulting in higher flux. In the case of constant feed side pressure (which is generally atmospheric pressure), permeate side pressure determines the permeation rate and because of the temperature dependency of permeant vapor pressure, so does feed temperature. The relative liquid transfer depth in equation 3 will be extended as permeate side pressure increases and it will be maximal ($\delta = \delta_L$) when $P_2 = P^*$. In this case, mass transfer will occur entirely as liquid permeation and no phase change will occur within the membrane.

3. MATERIALS AND METHODS

A. Experimental apparatus

The schematic diagram of the experimental apparatus is shown in Figure 2. A 2 %w/w aqueous solution of ethanol was pumped from a reservoir to the shell side of a 10 μm thick 100 cm^2 polydimethylsiloxane (PDMS) membrane (GKSS Research Institute, Geesthacht, Germany) housed in a stainless steel SEPA CF module (Osmonics Inc. Minnetonka, MN, USA). A negative pressure gradient was created by a vacuum pump (model GEM-8890, Welch Vacuum Technology Inc., Skokie, Illinois) maintained by a regulator (model 1421, Welch Vacuum Technology Inc. Skokie, Illinois) and monitored by a vacuum barocel gauge (Type 658, Edwards, Wilmington, USA) with a precision of 0.01 mmHg . The vapor phase permeate was connected via a three-way valve to one of two parallel series of two cold-finger condenser traps immersed in liquid nitrogen baths, alternating between each series at the hourly sampling intervals to allow continuous operation of the system while emptying the trap contents.

B. Sample analysis

The collected frozen permeate was melted and weighed. One mlit of each sample was diluted ten-fold while the remaining sample was returned to the feed tank to maintain constant initial ethanol concentration. Ethanol concentrations in the diluted permeates were analyzed in triplicate using a gas chromatograph (model HP 5890 with FID detector, Hewlett-Packard, Texas) equipped with a DB-WAX 20M column 30m long and 0.25mm diameter [32]. Injection and detector temperatures were both 170 $^\circ\text{C}$ and oven temperature was 35 $^\circ\text{C}$. Total flux was calculated using the following equation:

$$\text{Flux}_{\text{total}} = \frac{Q}{S \cdot t} \quad (4)$$

where Q , t and S are permeate quantity, pervaporation time interval and effective membrane surface area, respectively. Ethanol selectivity was calculated using

equation 1.

Total flux and ethanol selectivity were measured for Reynolds numbers of 500, 1000, 1500, 2000, 2200 and 2500, feed temperatures of 30, 40, 50 and 60°C and

permeate-side pressures of 1, 10, 20 and 40 mmHg. The effects of operating parameters on total flux and ethanol selectivity were evaluated by analysis of variance. All experimental conditions were repeated three times.

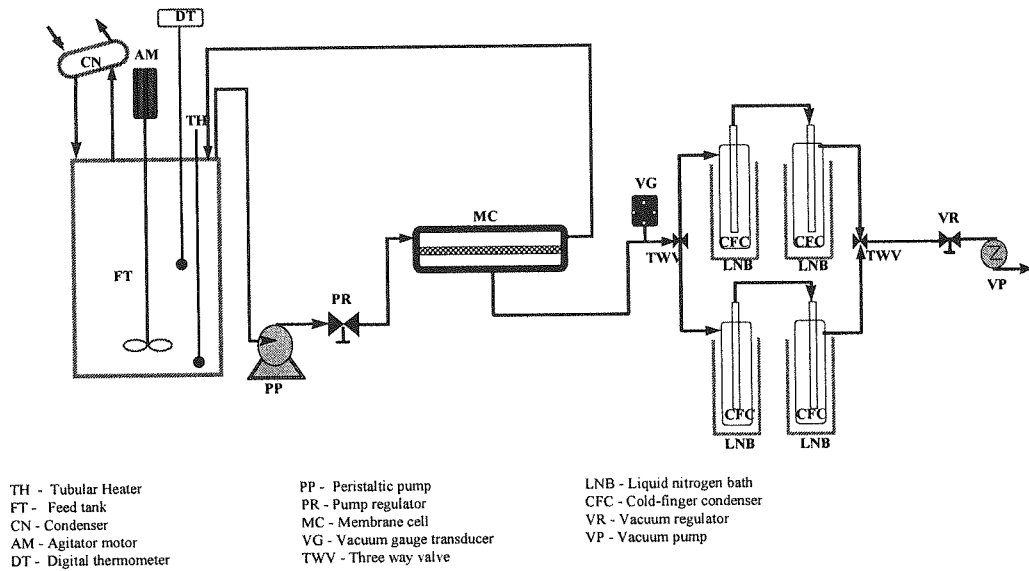


Figure 2: Schematic diagram of the experimental apparatus.

Steady state permeation was reached in fourth hours at all feed temperatures (Figure 3a), suggesting that both the concentration layer on the inside membrane surface (boundary or polarization layer) and the resistance of the thin membrane to passing permeate were low.

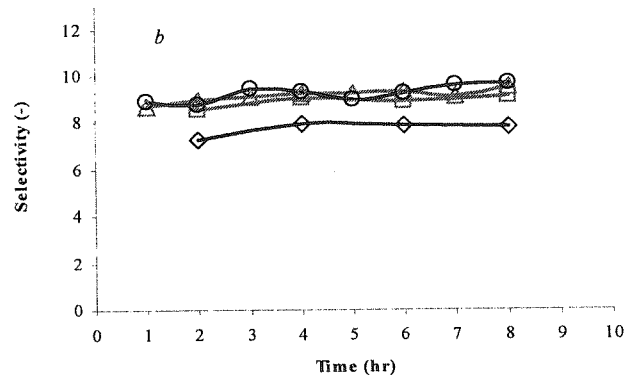
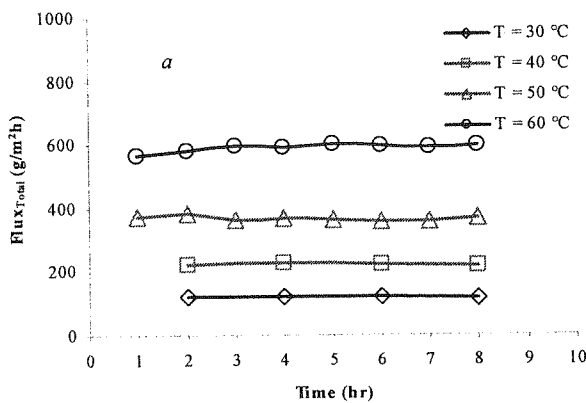


Figure 3: Time dependency of total flux (a) and ethanol selectivity (b) at different feed temperatures ($C_0 = 2\%w/w$, $P_2 = 1\text{ mmHg}$, $Re = 1000$).

C. Effect of volumetric flow rate

The effect of Reynolds number on total flux and ethanol selectivity at different feed temperatures for a fixed permeate-side pressure of 1 mmHg are shown in Figure 4. Flow regimes into a channel are laminar, transitional or turbulent for $Re < 500$, $500 < Re < 2000$ and $Re > 2000$, respectively [33].

Total flux and ethanol selectivity increased slightly versus Reynolds numbers of 500 to 2500 at all feed temperatures. Total flux increased by about 5% while the effect on selectivity was an increase of about 3% at 60°C. Flux and ethanol selectivity values were consistently higher at 60°C, although marginally in the case of selectivity. Feed volumetric flow rate thus had minimal

impact on ethanol enrichment in the permeate. It may thus be inferred that little or no polarization layer formed even at lower feed temperatures on the membrane surface.

D. Effect of permeate-side pressure

Total flux and ethanol selectivity versus permeate-side pressure are plotted at different feed temperatures in Figure 5. Total flux curves showed a non-linear decrease at all temperatures as permeate-side pressure was increased. Due to reduced driving force (permeate-side pressure), permeation rate dropped and approached zero as permeate-side pressure approached permeant vapor pressure. Vapor pressures of pure water and ethanol at 30°C are 32 and 74 mmHg, respectively. The vapor pressure of a 2 %w/w ethanol-water solution, calculated by Raoult's law, is 33 mmHg at 30°C and 57, 95 and 153 mmHg at 40, 50 and 60°C, respectively. Permeation rate should therefore approach zero as permeate-side pressure approaches the solution vapor pressure. However, extrapolating the total flux curves in Figure 5 gives permeate-side pressures of 28, 43, 48 and 57 mmHg for feed temperatures of 30, 40, 50 and 60°C, respectively at zero total flux. These differences, which are greater at higher feed temperatures, may be due to molecular interactions between ethanol and water and the membrane.

Permeate-side pressure also affects the pressure profile as well as the liquid transfer depth within the membrane, lower pressure inducing a higher pressure gradient within the membrane and decreasing the liquid transfer depth. As permeate-side pressure increases, the liquid transfer depth penetrates into the vapor transfer space and mass transfer becomes dominated by the slower liquid transfer, decreasing the total permeation rate. When the permeate-side pressure is held near the solution vapor pressure, the membrane is nearly filled with liquid, decreasing the permeation rate to zero. But as noted above, this occurred at permeate-side pressure levels lower than solution vapor pressures probably because of molecular interactions between components.

It was also shown that different feed temperatures had little effect on selectivity while permeate-side pressure was held below 20 mmHg but significant effect at pressures above 20 mmHg, at which selectivity increased as much as four-fold. This suggests that more ethanol can pass through the membrane when the liquid transfer depth increases relative to the vapor transfer depth within the membrane. In other words, ethanol may pass more readily than water through the wet space inside the membrane. Its solubility parameter is closer to that of the membrane and it should therefore solve into the membrane molecular structure more easily than water does. Selectivity may therefore be determined by the liquid transfer depth and will be greater when permeate-side pressure approaches solution vapor pressure.

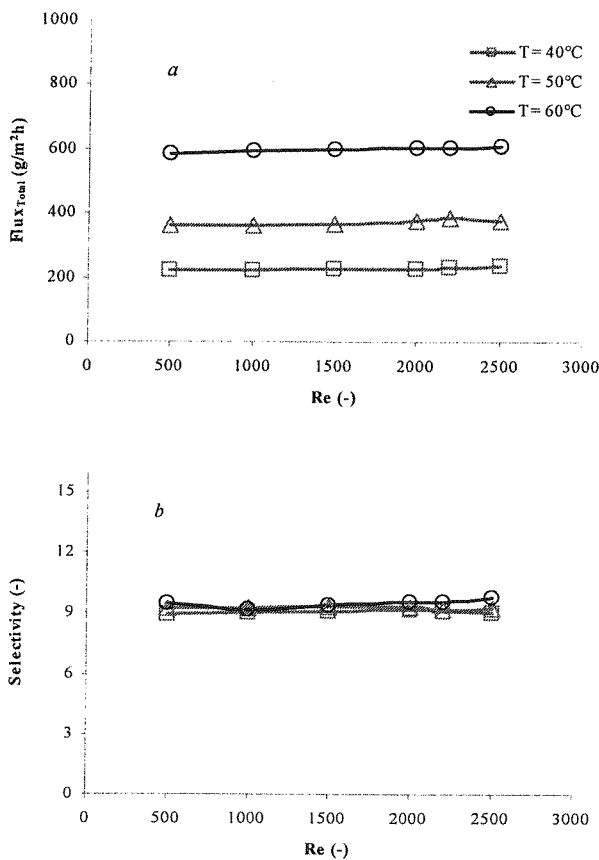


Figure 4: Effect of Reynolds number on total flux (a) and ethanol selectivity (b) at different feed temperatures ($C_0 = 2$ %w/w, $P_2 = 1$ mmHg).

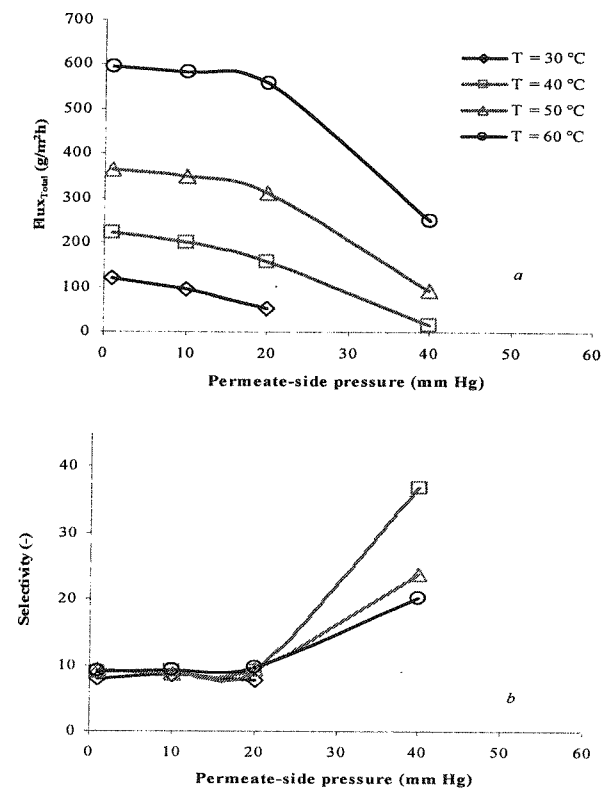


Figure 5: Effect of permeate-side pressure on total flux (a) and ethanol selectivity (b) at different feed temperatures ($C_0 = 2$ %w/w, $Re = 1000$).

E. Effect of feed temperature

Total flux and ethanol selectivity versus feed temperatures are plotted at different permeate-side pressures in Figure 6. Total flux increased significantly with increasing feed temperature, especially at the lower permeate-side pressures. The thin membrane likely reached a steady state temperature very quickly by heat transfer from the feed solution. At increased feed temperatures, segmental motion in the membrane polymer chains should increase, creating increased free space for diffusion of permeants within the membrane and hence an increased membrane diffusion coefficient, thereby increasing total flux.

Figure 6b shows that ethanol selectivity did not change significantly at different feed temperatures while the permeate-side pressure was held below 20 mmHg. But when permeate-side pressure was held at 40 mmHg, selectivity decreased significantly as feed temperature increased. The solution vapor pressure at a feed temperature of 40°C is very close to this pressure, thus filling the membrane with liquid and increasing selectivity. At feed temperatures of 50 and 60°C and a permeate-side pressure of 40 mmHg, solution vapor pressure increased and the liquid transfer depth decreased which partially explain why selectivity decreases beyond 40°C at

permeate-side pressure of 40 mmHg.

F. Statistical analysis of the effects of feed temperature and permeate-side pressure on the flux and selectivity

Flux and selectivity depended on both feed temperature and permeate-side pressure separately and in combination (Figures 5 and 6). Total flux and ethanol selectivity at each feed temperature did not change significantly while permeate-side pressure was below 20 mmHg. But at 40 mmHg, closer to the vapor pressure of the permeants, high variation of flux and selectivity was observed. At this permeate-side pressure, no flux was obtained for a feed temperature of 30°C whereas at feed temperatures of 40, 50 and 60°C, flux was 15.85, 92.67 and 251 g/m²h and selectivity 36.87, 23.79 and 20.20, respectively. Analysis of variance for total flux and ethanol selectivity is given in Table 1. The plan was a 4×4 factorial experiment (four feed temperatures and four permeate-side pressure levels) and three repetitions for each treatment combination. Feed temperature and permeate-side pressure had a highly significant effect on total flux, but the effect of permeate-side pressure on selectivity was greater than that of feed temperature.

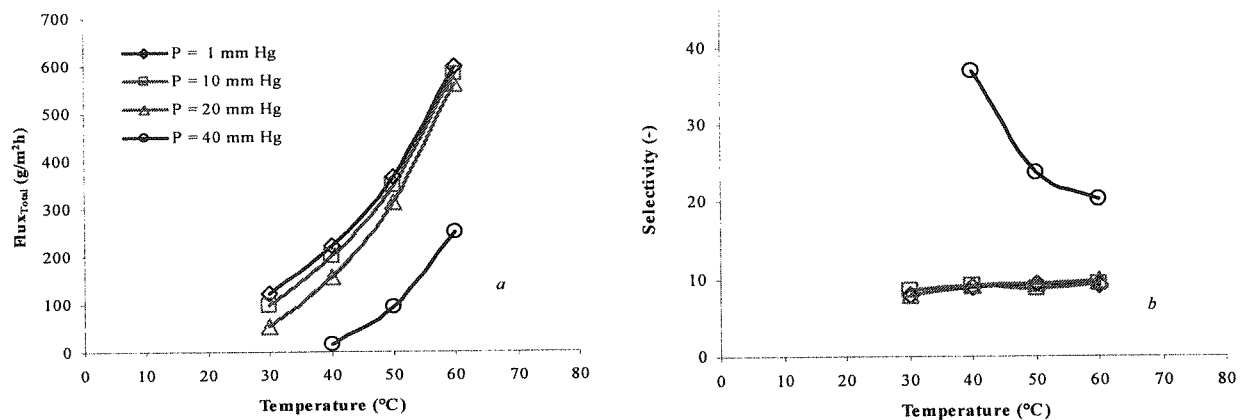


Figure 6: Effect of feed temperatures on total flux (a) and ethanol selectivity (b) at different permeate-side pressures ($C_0 = 2\%w/w$, $Re = 1000$).

TABLE 1:
ANALYSIS OF VARIANCE FOR TOTAL FLUX AND ETHANOL SELECTIVITY

Source of variance		Degrees of freedom	Sum of square	Mean square	Computed F	Probability
Total flux	Repetition (R)	2	97	49	2.45	0.1060
	Pressure (P)	3	350287	116762	5877	0.0001
	Temperature (T)	3	1237300	412433	20761	0.0001
	T*P	8	22589	2824	142	0.0001
	Error	26	516	20	-	-
Ethanol selectivity	Repetition (R)	2	0.46	0.23	0.83	0.4470
	Pressure (P)	3	1696	565	2035	0.0001
	Temperature (T)	3	106	35.2	127	0.0001
	T*P	8	283	35.4	127	0.0001
	Error	26	7.22	0.28	-	-

4. CONCLUSIONS

Pervaporation may be a useful technique to separate ethanol from dilute ethanol-water mixture. The following conclusions can be drawn from this work:

Total flux and ethanol selectivity did not change significantly versus Reynolds number at different feed temperatures, but their variations versus Reynolds numbers of 500 to 2500 were about 5% and 3%, respectively.

Total flux decreases non-linearly as permeate-side pressure increases. Ethanol selectivity does not change significantly at permeate-side pressures below 20 mmHg, but increases at permeate-side pressures above 20 mmHg.

Total flux significantly increases as feed temperature increases. Selectivity does not increase significantly at permeate-side pressures below 20 mmHg, but decreases at pressures above 20 mmHg.

Analysis of variance determined that permeate-side pressure, feed temperature and feed temperature-permeate-side pressure interaction all have significant effects on both total flux and ethanol selectivity.

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