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Pervaporation of Aqueous Ethanol Mixtures through Poly(Dimethyl Siloxane) Membranes

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Abstract

The separation of ethanol/water mixtures by pervaporation with a poly(dimethyl siloxane) membrane has been studied. The membrane exhibited ethanol selectivity during all process runs. Investigations focused on the effects of temperature and permeate-side pressure on membrane transport with dilute ethanol feed solutions. An increase in temperature increased the flux exponentially but had little effect on selectivity. As the permeate-side pressure was increased, the flux decreased. Selectivity did not change appreciably over the pressure range evaluated. Studies also analyzed the effect of feed concentration on flux and selectivity. Flux increased and selectivity decreased as the ethanol feed concentration increased. The permeate concentration profile is superior to a standard vapor–liquid equilibrium curve at low ethanol feed concentrations.

INTRODUCTION

Pervaporation describes a membrane unit operation where a liquid mixture is separated by partly vaporizing it through a semipermeable membrane barrier. Therefore, pervaporation differs from the other members of the membrane family, i.e., reverse osmosis, ultrafiltration, microfiltration, gas permeation, and electrodialysis, because a phase change occurs during the separation. Pervaporation holds a great potential for utilization by the traditional chemical industry and in emerging areas such as environmental and biochemical engineering. Pervaporation is very low on the relative scale of technical maturity and industrial process utilization.

In a simple pervaporation system the liquid feed flows across the membrane surface (or into the membrane module) normally at or near ambient conditions of pressure and temperature. Retentate and permeate streams are produced, similar to other membrane unit operations. The membrane swells from contact with the feed solution. Its permselectivity is due to the affinities of the feed compounds for the swelled layer and their varying mobilities in it.

Mass transfer in pervaporation, like reverse osmosis, can be considered to occur by a solution-diffusion mechanism, although other ways of describing the behavior have been presented. The permeation characteristics of the membrane are therefore highly dependent on the solubility and diffusivity characteristics of the system. Pervaporative transport has three basic steps; i) selective sorption into the membrane on the feed side, ii) selective diffusion through the membrane, and iii) desorption into the vapor phase on the permeate side (*1*).

Transport in the diffusional step in the mechanism can be easily described by Fickian diffusion.

$$J_i = D_i \frac{dc_i}{dl} \tag{1}$$

where J_i is the flux of Component i, D_i is the diffusion coefficient, c_i is the concentration of Component i in the membrane, and l is the perpendicular distance into the membrane surface. The interfacial equilibrium for sorption and desorption can be expressed as

$$c_i = K_{ci} a_i \tag{2}$$

where K_{ci} is the sorption coefficient and a_i is the component activity.

The driving force is the chemical potential gradient across the membrane. This can be related to the transmembrane pressure driving force. The flux of any component through the membrane can be described by the equation below which assumes constant diffusivity.

$$J_i = \frac{D_i K_{ci}}{l} \left[\gamma_i x_i - \frac{y_i P_p}{P_i} \right] \tag{3}$$

where x_i is the feed concentration of Component i, y_i is the composition of Component i in the permeate, γ_i is the activity coefficient for Component i, l is the membrane thickness, P_p is the permeate-side pressure, and P_i is the saturation pressure of Component i.

A representation of pervaporative transport for a range of concentrations requires a concentration-dependent diffusivity term. The single component diffusivity can be represented by an exponential:

$$D_i = D_{i0} e^{A_i c_i} \tag{4}$$

where D_{i0} is the diffusion coefficient at infinite dilution and A_i is a plasticization constant.

Substitution of Eqs. (4) and (2) into Eq. (1) and subsequent integration yields an expression for individual component transport through the membrane for the concentration dependent case.

$$J_i = \frac{D_{i0}}{A_i l}\left[e^{A_i K_{ci}} - e^{A_i K_{ci}(P_p/P_i)}\right] \tag{5}$$

The permeation rate for the separation of a binary mixture is composed of the fluxes of the desired and undesired permeating species. The membrane's selectivity can be expressed by a selectivity α.

$$\alpha = \frac{y_1/y_2}{x_1/x_2} \tag{6}$$

where x_1 = mass fraction of Component 1 in the feed
x_2 = mass fraction of Component 2 in the feed
y_1 = mass fraction of Component 1 in the permeate
y_2 = mass fraction of Component 2 in the permeate

This equation is for selective permeation of Component 1 and is therefore sometimes written α_1 or $\alpha_{1/2}$. A value greater than unity indicates selective permeation of "1" over "2" and a value less than unity indicates selective permeation of "2"over "1." Another form of selectivity, β, is less widely utilized in the pervaporation literature.

$$\beta = y_1/x_1 \tag{7}$$

This expression again relates to preferential permeation of Component 1 from a binary mixture.

Because of the phase change, the energy needed to obtain the separation is equivalent to the heat of vaporization of the permeate. The energy is usually difficult to recover, and maintenance of permeate cell temperature adds to the cost of operation. This is only a minor inconvenience compared to the high costs of azeotropic distillation and other complex separation methods. Since the permeating components are in the vapor state, they must be recovered. This is normally done by using conventional condensation techniques with coolants such as liquid nitrogen.

Neel (*2*) categorized several types of pervaporation operation. Some of these are really more process hybrids than process variations. The most widely utilized pervaporation process, vacuum pervaporation, employs a continuously applied vacuum on the downstream, permeate side. In sweeping gas pervaporation an inert gas stream passing the membrane on the permeate side continuously removes the permeating components. Pressure-controlled pervaporation maintains the permeate-side pressure at an optimal value for flux and selectivity.

Thermopervaporation utilizes the driving force of temperature difference between the liquid feed and walls of the permeate side of the unit. This process variant is initiated by vacuum pervaporation, which is then stopped, allowing the temperature gradient to take over. The permeate condenses on the cooled walls of the permeate-side cell or tubing. Another form of pervaporation utilizes increased pressure on the feed side while maintaining reduced or vacuum conditions on the permeate side. The process can increasingly become one of vacuum-aided reverse osmosis as the upstream feed pressure is increased at constant vacuum pressure on the downstream side.

Saturated vapor permeation is a form of pervaporation where the feed mixture is in the vapor phase. Extractive pervaporation is somewhat similar to extractive distillation and liquid–liquid extraction where a third component is introduced into the binary mixture to aid in the separation. In this operation the extracting and stripping concurrently occur on opposite sides of the membrane, providing the driving force. Pertraction employs a solvent on the permeate side of the process to maintain a lower activity and thereby allow permeation. No phase change occurs in this membrane unit operation.

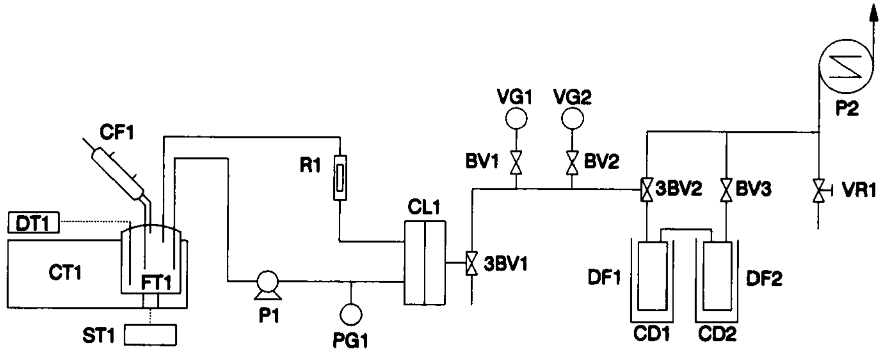
Pervaporation can be applied to a variety of separations and has potential for applications in many industries. In the chemical industry, pervaporation has been used to the greatest extent in the separation of organic/water mixtures. Systems for dewatering aqueous mixtures, e.g., dehydration of ethanol, isopropanol, and other simple alcohols, have reached commercial scale (*3*). In particular, commercial plants for the dehydration of ethanol/water mixtures and azeotropic separation have been demonstrated (*4*, *5*). These cases involve selective water permeation from low water concentration feed mixtures. This has a major commercial significance because of the complexity and expense of azeotropic and extractive distillation (*6*). In these cases, cellulose acetate and poly(vinyl alcohol) membranes have been successfully employed.

Selective organic permeation from aqueous feed mixtures, which is of interest in this research, is also possible upon correct membrane selection. The most popular membranes used in this application are silicone-based polymers, but much research is underway on other materials, e.g., poly(trimethylsilyl propyne) (PTMSP). In the area of biochemical engineering/biotechnology, pervaporation has been shown to be effective for alcohol recovery from fermentation broths (*7–12*). In the food and beverage industry, regulation of alcohol content in beer and wine production is another promising commercial application (*13*). In the environmental engineering fields of water purification, industrial and hazardous wastewater treatment, pervaporation can be utilized to remove trace organics, many of which are carcinogens (*14–17*).

Separation of isomeric compounds, e.g., the xylenes, or other organic/organic mixtures with similar chemical characteristics have been demonstrated on a laboratory scale, but large-scale commercialization has not been thoroughly examined (*18–22*). Regardless of what the azeotropic concentration is or whether the system is aqueous/organic or organic/organic, pervaporation can be applied, although more successfully to some cases than to others.

EXPERIMENTAL

The pervaporation system used in the research is a bench-scale, pressure-controlled unit shown in Fig. 1. Feed solution is continuously pumped to the feed port of the membrane test cell, flows across the membrane surface, and is removed from the cell via the retentate port. Permeate is collected as product by using two cold finger condensers placed in series and chilled by liquid nitrogen.



CT1 - Constant Temp. Bath
CF1 - Condenser (optional)
FT1 - Feed Tank
ST1 - Stirring Unit
DT1 - Digital Thermometer

P1 - Feed Pump
R1 - Feed Rotameter
CL1 - Membrane Cell
3BV1,2 - 3-Way Ball Valves
BV1,2,3 - 3 Way Ball Valves

VG1 - McLeod Vacuum Gauge
VG2 - Bennert Vacuum Gauge
CD1,2 - Cold-finger Condensers
DF1,2 - Dewars (for LIN)
VR1 - Vacuum Regulator
P2 - Vacuum Pump

FIG. 1. Pervaporation system process diagram.

The experimental studies utilized silicone membranes obtained from the General Electric (GE) Corporation, Membrane Products Operation, Medical Systems Division in Schenectady, New York. The membranes were symmetric poly(dimethyl siloxane) (PDMS), 25 μm in thickness, mounted to a polypropylene backing.

The feed was kept at constant temperature by having the feed vessel immersed in a constant temperature bath. The majority of the experiments were conducted at 30°C. The feed rate was maintained at 1500 cm^3/min. Since the single-pass recovery of the membrane module is very low, the concentration of the retentate is not that different from the feed. The membrane test cell consists of two 5 in. o.d. stainless steel, circular cell flanges bolted together and sealed by two interior gasket rings. The effective membrane area for permeation is 28.7 cm^2. Provisions were made to maintain the cell temperature, but as a result of the relatively low permeate rate through the membrane, the temperature drop between the feed and permeate streams due to the vaporization of the liquid feed was negligible.

The system's pressure was maintained by a direct drive vacuum pump capable of obtaining a vacuum of 1×10^{-4} torr. A vacuum regulator was utilized to control the permeate-side downstream pressure within ±1 torr. To measure the permeate-side pressure, a McLeod gauge was placed inline downstream from the pervaporation cell. It was accurate in the range 5 to 5×10^{-3} torr. A Bennert manometer was used for monitoring pressures above 5 torr. Permeate flux was measured gravimetrically, and the concentration of the permeate was determined by the use of a refractometer.

All pervaporation runs utilized a benchmark condition to evaluate any change in membrane characteristics over processing time. The standard process parameters were feed concentration, 5.0% w/w ethanol; feed temperature, 30°C; and permeate-side (downstream) pressure, ~1 torr. Benchmark permeate-side pressure, although reported as ~1 torr, was typically in the range of 0.75 to 1.5 torr. The research group has shown that there is no significant difference in flux and selectivity from 1 to 2 torr, but it did everything possible to keep close to 1 torr.

Pervaporation process studies were performed in such a way as to rule out any trend in the time history of processing to coincide with the process parameter trends under investigation. Proper preconditioning of the membrane was performed when a new membrane was utilized and after process parameters were changed.

RESULTS AND DISCUSSION

The experimental studies focused on the effect of the feed temperature, permeate-side (downstream) pressure, and feed concentration on pervaporative transport. The data are quantified in terms of membrane flux and selectivity for the PDMS membrane.

The effect of temperature on permeate flux, using a 5% ethanol feed concentration at 1 torr, is presented in Fig. 2. As temperature is increased from 20 to 90°C, flux increased from 77.6 to 1906 g/m^2·h. The exponential trend in the data can be easily seen. The ethanol component flux increased from 24.8 to 591 g/m^2·h as the feed temperature increased from 20 to 90°C, respectively. The benchmark permeate flux obtained in the study at 30°C is 136 g/m^2·h. An excellent linear correlation exists between the log of the specific permeation rate and the reciprocal absolute temperature, showing that an Arrhenius-type relationship is quite valid. An activation energy of 9.4 kcal/mol was calculated. According to the permeation transport mechanism, the activation energy can be considered to be composed of two components, the energy for diffusion and the heat of sorption.

It appears that as the feed temperature increases, the concentration of ethanol in the permeate increases slightly to a maximum, then decreases (Fig. 3). The ethanol permeate concentration ranges from 31 to 35.5% over the temperatures studied. The resulting selectivities vary from 8.94 to 10.5. The maximum selectivity is observed at about 50°C, although this may not be truly significant. This phenomena may be due to the varying

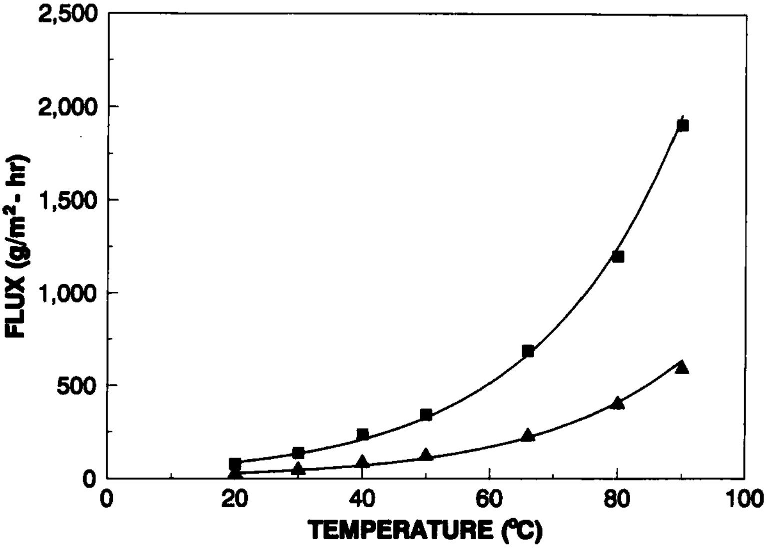


FIG. 2. Flux vs temperature for a 5% ethanol feed mixture at 1 torr downstream pressure: (■) total flux and (▲) ethanol flux.

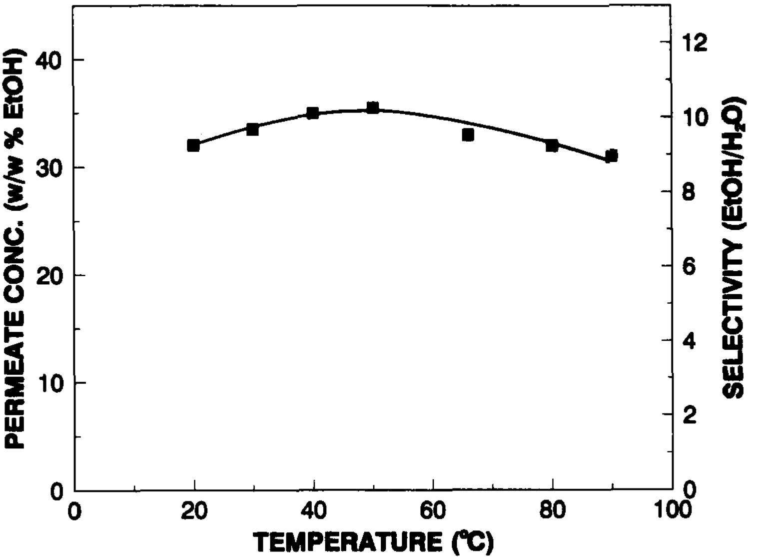


FIG. 3. Permeate concentration and ethanol selectivity vs temperature for a 5% ethanol feed mixture at 1 torr downstream pressure.

degrees of swelling occurring at the different temperatures coupled with the greater diffusion rates. The effect of temperature on selectivity is highly system (membrane/mixture) dependent. The rate governing step, sorption or diffusion, is a key factor which may change with the variable being studied.

The membrane is quite stable when operating at high temperatures. Extended time studies during previous runs showed that the membrane's performance characteristics of flux and selectivity are not affected by temperatures of 90°C. Several membranes were repeatedly subjected to long-term processing and temperature variation studies. In all cases the membranes were able to reproduce benchmark process values. The silicone-based polymers appear to be more thermally stable than other alcohol-selective membrane materials such as PTMSP.

The increase in temperature greatly improves system productivity with the PDMS membrane since the slight change in selectivity at high temperature is insignificant in comparison to the ethanol permeation rate. Some additional processing concerns when operating at high temperatures must be addressed. When the flux is greatly increased, additional heat must be supplied to the membrane module to provide the needed heat of vaporation for the permeate. If this is not done, the liquid stream and system will experience a cooling effect since this is the only available heat source.

The effect of permeate-side pressure on flux and selectivity was also evaluated by varying the downstream pressure from 1 to 30 torr. Feed conditions were kept constant at 5.0% ethanol and 30°C. When the permeate-side pressure was increased from 1 to 30 torr, the total flux dropped from 136 to 56.8 $g/m^2 \cdot h$ (Fig. 4). The ethanol flux was affected in a similar manner. The ethanol flux at 1 torr was 45.6 $g/m^2 \cdot h$ and decreased almost linearly to 17.0 $g/m^2 \cdot h$ at 30 torr. The pervaporative transport model previously described can explain this trend since an increase in the vapor pressure on the downstream side decreases the driving force. Even though a lower pressure increases productivity, it costs significantly more to use a high vacuum unit.

Permeate-side pressure did not seem to have a major effect on membrane selectivity for pressures less than 30 torr. Permeate concentration fluctuated from 34 to 30% ethanol representing a selectivity range of 9.78 to 8.14. The selectivity drops slightly at 30 torr and falls even further as the limiting pressure is approached.

The effect of the ethanol feed concentration on the pervaporation of the aqueous mixture was studied at a feed temperature of 30°C and a permeate-side pressure of 1 torr. The total flux increases as the concentration of ethanol in the feed increases (Fig. 5). At 0% ethanol the flux is 102 g/

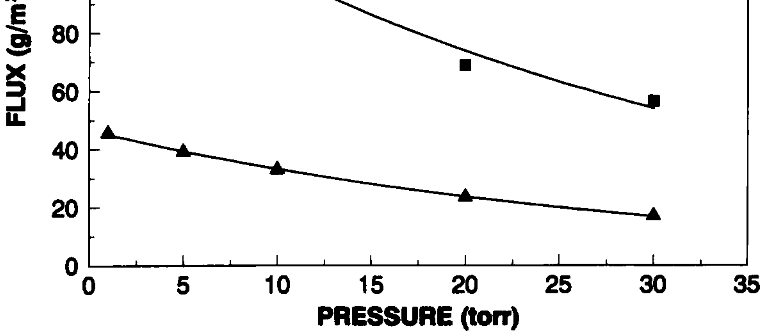


FIG. 4. Flux vs pressure for a 5% ethanol feed mixture at 30°C: (■) total flux and (▲) ethanol flux.

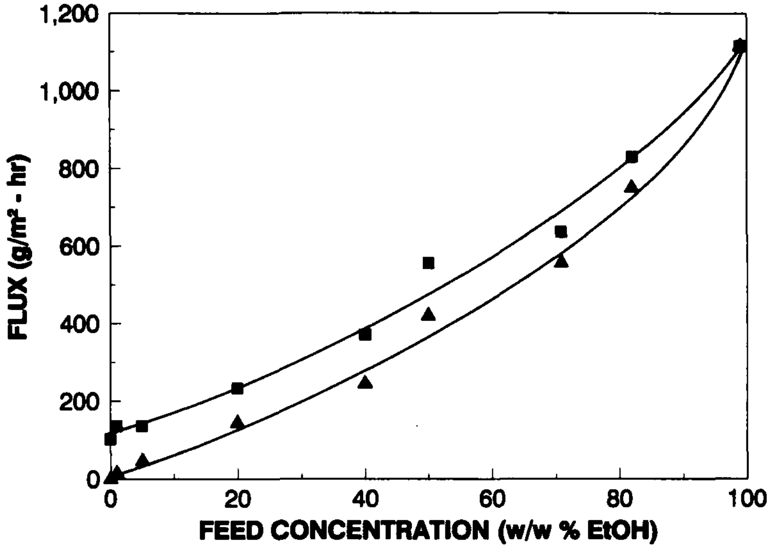


FIG. 5. Flux vs feed concentration at 30°C and 1 torr downstream pressure: (■) total flux and (▲) ethanol flux.

$m^2 \cdot h$, and at 100% ethanol the flux is 1115 $g/m^2 \cdot h$. The process trend appears to follow an exponential increase in flux with feed concentration. The ethanol flux also shows a correlation similar to that of the total flux.

The ethanol concentration of the permeate continually increases with the feed concentration, although the differential increase decreases. This is shown in the selectivity data where the selectivity decreases sharply from 0 to 50% ethanol, then approaches the unity asymptote (Fig. 6). This information, upon which a vapor–liquid equilibrium (VLE) curve at 760 torr and 30°C is superimposed (Fig. 7), shows that the commercial pervaporation potential for the PDMS membrane is limited to the low ethanol feed cases of less than 20%.

An examination of the effect of feed concentration on membrane performance shows that as the concentration of the more selective permeant increases, so does the flux and permeate concentration. This can be explained in terms of the dependency of the kinetic and thermodynamic parameters on concentration. As the ethanol feed concentration increases, the diffusion rate increases as does the membrane's solubility rate for the ethanol.

A comparison of the results with other silicone-based polymers is presented in Fig. 8. They show that a polymer blend of PDMS and PTMSP

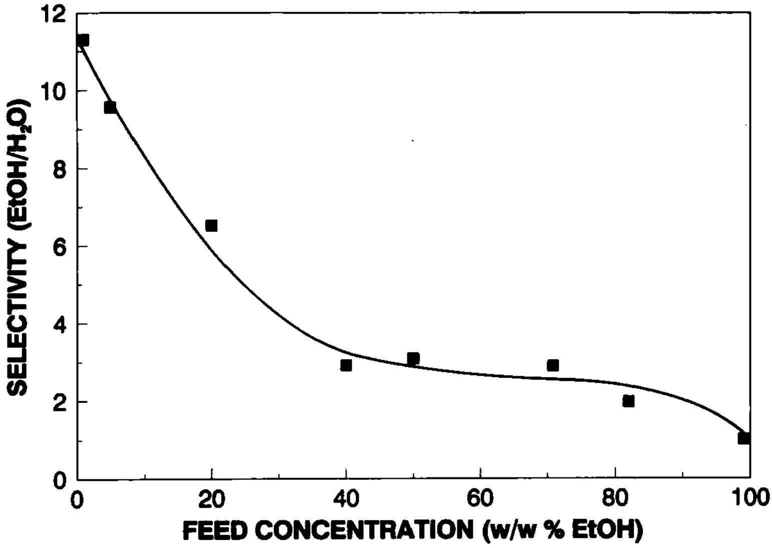


FIG. 6. Ethanol selectivity vs feed concentration at 30°C and 1 torr downstream pressure.

produces superior results to any of the others investigated. Nagase et al utilized a blend of 69% PDMS and 31% PTMSP (*23*). The results of Ishihara and Matsui with a PDMS membrane are also well above the standard VLE curve (*24*).

CONCLUSIONS

Pervaporation is a relatively new membrane technology with great potential for utilization in the traditional chemical industry as well for biochemical and environmental engineering. Analysis of selective organic permeation from aqueous systems pervaporation literature has shown that a majority of the papers list their potential application in biochemical separation. Most papers cite the need to create energy from renewable energy sources and list pervaporation as an effective way of removing product alcohols while also enhancing the fermentation process.

The parameters affecting the selective separation of ethanol through pervaporation membranes have been studied. A bench-scale system was used in conjunction with a poly(dimethyl siloxane) (PDMS) membrane. Three experimental studies consisting of the variation of temperature, downstream pressure, and feed concentration were investigated. These studies were designed to optimize pervaporation as well as to determine its limits.

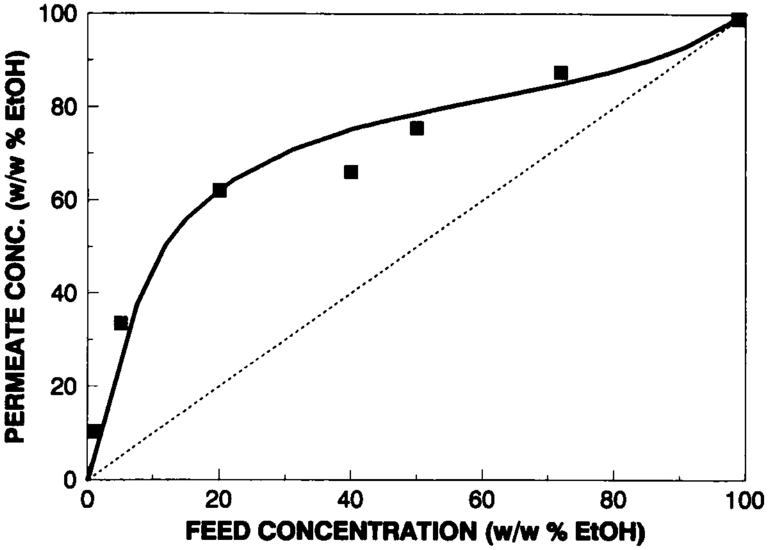


FIG. 7. Permeate concentration vs feed concentration at 30°C and 1 torr shown with standard vapor–liquid equilibrium (VLE) curve at 30°C: (■) pervaporation data and (——) VLE.

Variation of the flux with the process parameters was seen in all three studies. The flux increases with increasing temperature in an Arrhenius-like relationship. The pressure study showed a decrease in flux with increasing downstream pressure. The flux had a slight exponential increase with increasing feed concentration.

Selectivity was not greatly effected by temperature, and it was observed to increase slightly. The downstream pressure has little effect on selectivity in the range <30 torr. Although permeate concentration increases with increasing feed concentration, the selectivity decreases to unity. It has been shown that the membrane is most selective at lower feed concentrations.

Based on the data obtained for a 5% ethanol feed concentration, the optimal conditions for permeation occur at the highest temperature and lowest permeate pressure, 90°C and 1 torr, where the flux was 1906 $g/m^2 \cdot h$ and the selectivity was 9. The maximum selectivity for a 5% feed mixture appears to occur at approximately 50°C, yielding a value of 10.5.

The commercial application potential for this type of separation is biggest in environmental and biochemical engineering fields because of the low concentrations of solutes to be removed. Further research on this class of separations is warranted to determine a more thorough understanding of the process. Enhancing membrane performance, e.g., with novel polymer

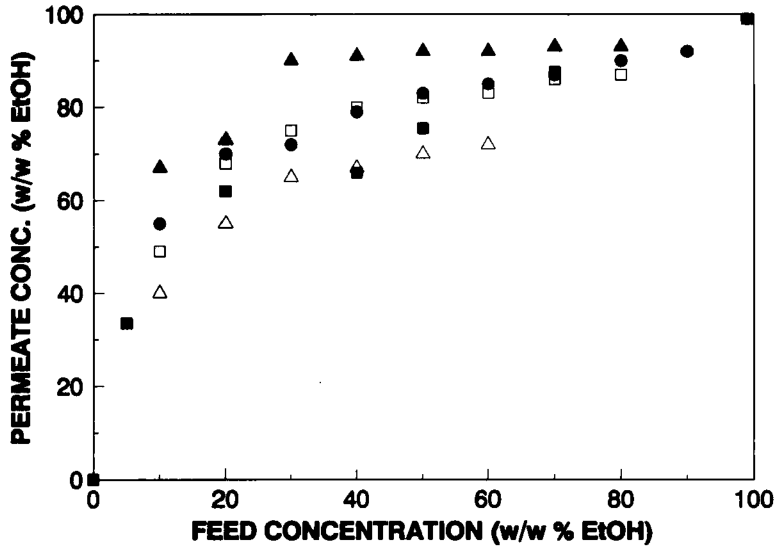


FIG. 8. Comparison of experimental data to other silicone-based membranes: (■) Slater et al.; (▲) Ref. *23*, 69% PDMS/31% PTMSP; (□) Ref. *23*, 55% PDMS/45% PTMSP; (△) Ref. *25;* and (●) Ref. *24*.

blends or surface modifications, is essential to make pervaporation a more viable and competitive commercial separation process.

SYMBOLS

a	component activity
A	plasticization coefficient
c	concentration in the membrane
D	diffusion coefficient
D_0	diffusion coefficient at infinite dilution
J	flux
K_c	sorption coefficient
l	membrane thickness
P	saturation pressure
P_p	permeate-side (downstream) pressure
x	concentration of the feed
y	concentration of the permeate
α	selectivity
β	enrichment factor
γ	activity coefficient

Subscripts

i denotes the individual component
1 Component 1
2 Component 2

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