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

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To cite this Article Slater, C. S. , Devine, K. M. and Meier, A. J.(1997) 'Pervaporation for the Separation of Methyl Ethyl Ketone from Binary Process Mixtures', Separation Science and Technology, 32: 6, 1127 — 1142
To link to this Article: DOI: 10.1080/01496399708000951
URL: http://dx.doi.org/10.1080/01496399708000951

SEPARATION SCIENCE AND TECHNOLOGY, 32(6), pp. 1127–1142, 1997

Pervaporation for the Separation of Methyl Ethyl Ketone from Binary Process Mixtures

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ABSTRACT

The separation of methyl ethyl ketone (MEK) from binary process mixtures containing water and ethanol has been examined. These studies demonstrate the capability of pervaporation membranes to effectively separate MEK from various process streams in industry. Extensive organic permeation studies were performed using silicone composite membranes to evaluate the effect of temperature, permeate-side pressure, and feed concentration on flux and selectivity. Two organophilic membranes, a high selectivity low flux membrane and a low selectivity high flux membrane, were compared. Dehydration of a MEK–water mixture was also effectively performed with a polyvinyl alcohol composite membrane. MEK was separated only slightly from an ethanol mixture using a silicone membrane.

INTRODUCTION

Pervaporation is a membrane-based separation method that shows much promise for the future as a viable means of separating mixtures in process streams. Pervaporation can be used to dehydrate organic solvents in order that they be recovered and reused. Pervaporation can also be used to selectively remove trace amounts of organics from process streams. The result of this process is twofold. First, if the organics are successfully removed from the aqueous mixture, the now purified water can be recycled or discharged without environmental consequences. Second, the newly recovered solvent can be further purified by a dehydration process and reused in the process stream as needed. Thus, the use of pervaporation can be looked upon as an environmentally and economically advantageous process (1).

The first successful commercialization of pervaporation technology was organic solvent dehydration. Commercial plants to dehydrate solvents such as isopropanol, acetone, ethanol, etc. are in worldwide operation (2–4). These systems use hydrophilic membranes to selectively permeate water and produce a "dry" organic retentate. Commercial pervaporation systems for organic removal from aqueous mixtures have been successfully applied in environmental, biotechnology, and food/beverage applications (3, 4). By far the most promising and challenging area for growth is the application of pervaporation technology to organic–organic separations common to the petrochemical and specialty chemicals industry. Pilot-scale separation of alcohols from methyl and ethyl *tert*-butyl ethers has been developed, and there exists the potential to apply this technology to many types of organic mixtures (3, 4).

Previous research by the authors has used organophilic pervaporation to separate a variety of organic–water mixtures (5–12). Recently a comparative study was performed to determine the relative permeability of various solvents used in specialty chemical and biochemical production (13). Some of these solvents included furfural, pyridine, ethyl acetate, a variety of alcohols, and others. The objective of the current work was to study methyl ethyl ketone (MEK) in detail and see how various process parameters affect the separation of dilute amounts of MEK from aqueous solutions. Also studied was dehydration of MEK solutions and the separation of an anhydrous mixture of MEK–ethanol.

Thorough presentations of the theory of pervaporation have been presented by these authors and others elsewhere (4, 14–16), and only the basic components as necessary to this paper are presented herein. The reader is referred to the above references for a further understanding of the various theoretical considerations on the subject. In the standard pervaporation process, a liquid stream is passed across a membrane at (or near) atmospheric pressure while a low pressure (close to vacuum) is maintained on the permeate side. A chemical potential gradient is established between the liquid feed side and the components that transport through the membrane and appear in the vapor phase on the permeate side. The low pressure can be initiated by use of a vacuum pump or other suitable device. The permeate-side pressure must be kept below the saturation vapor pressure of the permeants so that effective transport occurs. Pervaporation is typically differentiated from other membrane operations, such as reverse osmosis and gas permeation, since a phase change occurs.

The generally accepted transport theory used to describe the pervaporation process is the solution–diffusion mechanism. The three-step mechanism is:

1. Sorption into the membrane on the feed side
2. Diffusion through the membrane
3. Desorption from the membrane on the permeate side

The selectivity of the membrane is determined by the selective sorption and/or diffusion through the polymer. The desorption step is considered insignificant as a resistance to transport.

The flux of any component through the membrane can be expressed by

$$J_i = L_i(\Delta \mu_i / l) \tag{1}$$

where L_i is the phenomenological permeability coefficient, which is a function of both solubility and diffusivity of the permeants in the membrane, $\Delta \mu_i$ is the chemical potential difference across the membrane, and l is the membrane thickness. Total flux is easily determined from experimental data knowing total mass collected per membrane area in a given period of time. Component flux is determined based on the permeate composition. Selectivity for binary mixture separation is expressed as

$$\alpha_A = (y_A/y_B)/(x_A/x_B) \tag{2}$$

where y_A, x_A = mass fraction of component A in the permeate (y) and feed (x), respectively
y_B, x_B = mass fraction of component B in the permeate (y) and feed (x), respectively

The use of pervaporation technology to separate MEK mixtures has been investigated by several researchers. Baker and coworkers (17) looked at the separation of ethanol from MEK solutions using various types of membranes. Selectivity and flux were compared for 24 μm Elvamide, polyetherimide/PDMS/Elvamide membrane samples, and polyetherimide/crosslinked Elvamide membrane modules. Their results indicate that the azeotrope is effectively broken.

Behling et al. (18) investigated the application of membrane vapor permeation for the separation of hydrocarbons from waste vapor streams. MEK was one of the solvents examined in the study. The authors indicated that pressure had a significant effect on vapor permeation. As pressure increased, the flux increased rapidly, demonstrating the strong pressure dependence that exists between organic vapors and solvent diffusion.

Pasternak et al. conducted detailed studies for Texaco (19, 20) whereby water was separated from MEK by use of pervaporation. In one of these studies (19), a dilute solution of MEK was concentrated by pervaporation through a fluorinated ion-exchange membrane. The advantages of Texaco's new technology is demonstrated in the separation of oxygen-containing compounds from water. Through their studies they were able to enrich streams containing trace amounts of MEK.

In their second study (20), water was separated from an MEK–toluene ternary solution dewaxing solvent using a polyvinyl alcohol composite on a polyacrylonitrile support. Their studies indicated that the membrane is effective in removing water from mixtures of hydrocarbons and organic oxygenates.

Zhou et al. performed MEK dehydration studies using electrochemically synthesized polypyrrole membranes (21). Their studies yielded promising results in both flux and selectivity.

MATERIALS AND METHODS

The membranes used for the majority of the organic permeation experimental studies were GFT-type 1170 silicone composite membranes provided by Carbone of America. This membrane is a "silicalite-filled" membrane, that is, composed of a ~20-μm layer of poly(dimethyl siloxane), PDMS, with hydrophobic inorganic materials, i.e., zeolites, incorporated into this layer to increase the membrane's selectivity toward organics. The percentage of zeolites in the 1170 is 60% by weight. Other types of membranes were also studied during the course of the investigations. One such membrane was a GFT-type 1160 membrane which does not have the "silicalite filling" and as such is a standard silicone composite. The thickness of the silicone layer is ~8 μm. A poly[1-(trimethylsilyl)-1-propyne], PTMSP, membrane with an asymmetric structure (~2 μm) was also investigated. Dehydration studies were carried out with a GFT-type 1000 polyvinyl alcohol (PVA) composite membrane, also supplied by Carbone of America. This membrane consists of a ~3-μm PVA selective layer on a polyacrylonitrile (PAN) support.

The experimental systems used was a Zenon bench scale pervaporation system (22) manufactured by Zenon Environmental, Inc. (Fig. 1). This system is composed of a 7-L (maximum) capacity feed tank and a membrane test cell that accommodates small rectangular membrane sheets yielding an effective area for transport of 7.91×10^{-3} m^2. Feed flows across the membrane in a true crossflow pattern. The unit has feed temperature and flow rate controls along with permeate-side pressure control. The vaporous permeate is collected in condensers submerged in Dewar flasks filled with liquid nitrogen. Permeate quantity was measured gravimetrically, and composition was determined through use of a refractometer. Benchmark conditions for the organic permeation studies were a feed

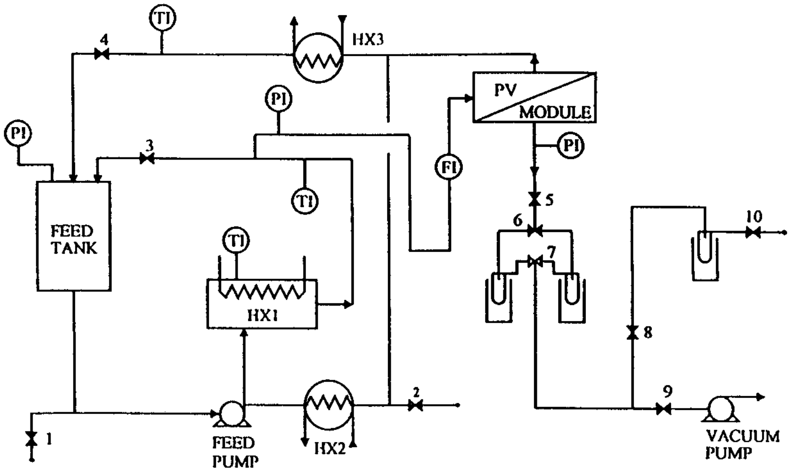

FIG. 1 Zenon bench-scale pervaporation system process schematic. Process valves (1–10), pressure indicators (PI), heat exchangers (HX1–3), temperature indicators (TI), and flow indicator (FI).

temperature of 50°C, MEK composition of 2.1% (by weight), permeate-side pressure of ~2 mbar, and feed flow rate of 9 L/min. The cell geometry and flow conditions correspond to a Reynolds number of 9480.

Experimental data were collected following a standard experimental protocol with a series of values collected during a run at prescribed conditions. The membrane was operated at the desired temperature, pressure, and hydrodynamic conditions for approximately 1 hour. This was found sufficient for steady state to be reached with the type of composite membranes used in this research. The first experimental value of the run is known as the "start-up" data, and the values obtained are not entered into the run's average. Typically 3 to 5 measurements are obtained under the same operating conditions by sampling at 30–60 minute intervals during the run. When there is a significant deviation in the samples from a given run in either permeate concentration or flux, additional runs are performed. Typical experimental error representative of other studies is shown for the temperature study at 50°C which yielded 5 data sets (permeate concentration and flux). The mean concentration was 76.8% and the range of experimental values was from 74.3 to 77.9 with a standard deviation of 1.45. The experimental flux data averaged 0.397 kg/m^2·h with a range of 0.388 to 0.412 and a standard deviation of 0.0103.

RESULTS AND DISCUSSION

MEK–Water Organic Permeation Study

The effect of feed concentration on the performance of the membrane system was studied at the benchmark conditions of 50°C and a permeate side pressure of 2 mbar using the GFT 1170 silicone composite membrane. Feed MEK concentration was varied from 1 to 15% (all concentrations are in weight percent). At a benchmark concentration of 2.1%, the total flux was 0.397 kg/m^2·h and the permeate concentration was 76.8%. This yields an organic selectivity of 162.3.

As the concentration of MEK in the feed increased, so did the flux of the MEK (Fig. 2). This demonstrates that as the concentration of the more permeable component increases, so does its corresponding flux. Total flux also increased with feed concentration. The water flux through the membrane remained relatively constant with increasing feed concentration (Fig. 2). Its average value was 0.092 kg/m^2·h, and this result indicated that MEK did not have a significant interaction with water transport in the process range studied. The effect of feed concentration on permeate concentration is basically indicated by the relationship between the component fluxes. Permeate concentration increases rapidly in the low feed concentration range, up to ~5%, and then levels off to approximately 89% at feed concentrations above 5% (Fig. 3). This value is above the standard vapor–liquid equilibrium (VLE) curve (Fig. 4). The membrane's selectivity to the organic is found to decrease with increase feed concentration because the permeate concentration only gradually increases after the 5%

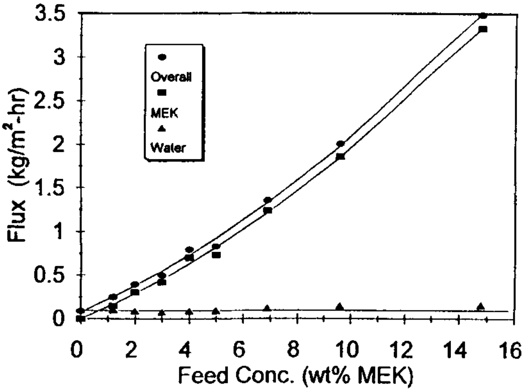

FIG. 2 Flux vs feed MEK concentration using the filled silicone composite membrane operating at 50°C and 2 mbar.

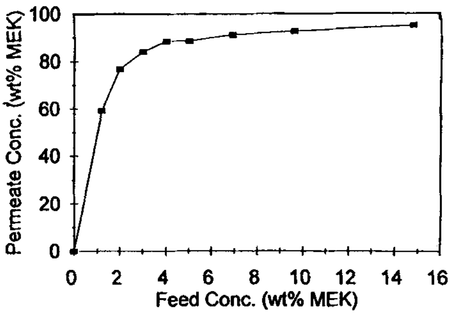

FIG. 3 Permeate MEK concentration vs feed MEK concentration using the filled silicone composite membrane operating at 50°C and 2 mbar.

feed concentration is obtained. The data in the 1 to 15% range were modeled and found to follow the following two quadratic equations:

$$J_{\text{total}} = 63.67x^2 + 13.67x + 0.0806 \quad (3)$$

$$J_{\text{MEK}} = 63.32x^2 + 13.19x \quad (4)$$

where x is the mass fraction of MEK in the feed, and the flux is in units of kg/m^2·h.

Temperature studies were performed at benchmark conditions as well. The results support the general trend that has been reported by these

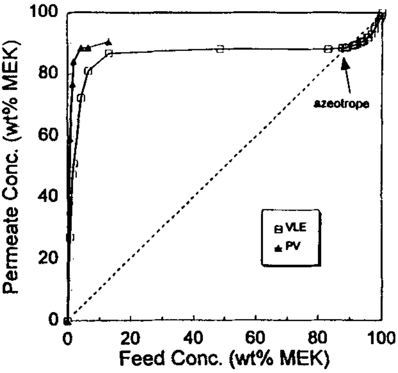

FIG. 4 Comparison of pervaporation "operating curve" with filled silicone composite membrane vs standard vapor liquid equilibrium (VLE) data. Experimental conditions at 50°C and 2 mbar. Experimental data shown.

authors and others with various organics, that flux increases with feed temperature. Temperature was varied over the range of 30 to 75°C and total flux increased from 0.225 to 1.04 kg/m^2·h. Both organic and water fluxes increased over the range studied (Fig. 5). Permeate concentration remained within the range 65.5 to 82.3%, appearing to drop slightly at higher temperatures. An Arrhenius-type plot of natural log flux versus the reciprocal of absolute temperature was obtained (Fig. 6), and the resulting equations for this relationship were determined:

$$J_{\text{total}} = 31943e^{-3616/T} \tag{5}$$

$$J_{\text{MEK}} = 4838e^{-3096/T} \tag{6}$$

$$J_{\text{H}_2\text{O}} = 938652e^{-5178/T} \tag{7}$$

where temperature is in degrees Kelvin and flux is in kg/m^2·h.

Pressure studies performed show how varying the permeate-side pressure affects the flux and selectivity. Flux dropped as permeate-side pressure was increased, and its rate of flux decline increased as pressure was increased (Fig. 7). Flux does not change significantly below 40 mbar. This would be of interest in operating commercial systems that are typically run at pressures much higher than the low range most researchers utilize. The permeate MEK concentration increases and was reported to be 98% at 75 mbar, since the water flux had dropped dramatically. MEK would continue to transport as pressure is increased, and its pure saturation vapor pressure is 333 mbar.

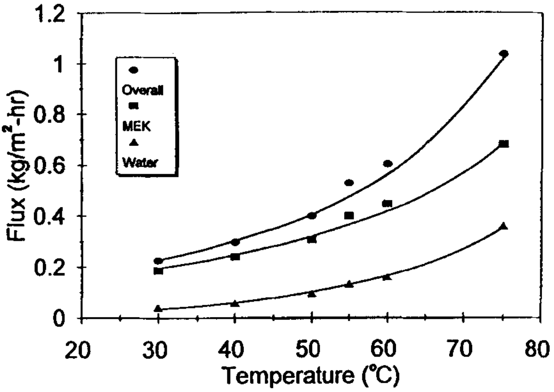

FIG. 5 Flux vs feed temperature using the filled silicone composite membrane operating at 50°C and 2 mbar.

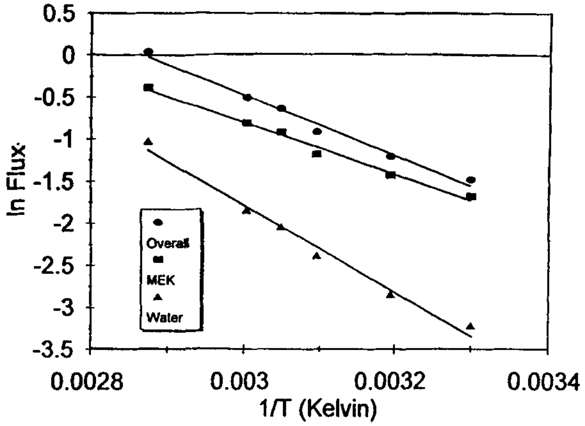

FIG. 6 Natural log, ln, of flux vs reciprocal of absolute temperature using the filled silicone composite membrane for a 2.1% (weight) MEK feed mixture and 2 mbar.

The effect of Reynolds number on separation was studied. The experimental conditions used for this phase of the research were the standard benchmark conditions: 2.1% MEK feed concentration, 50°C feed temperature, and a permeate-side pressure of 2 mbar. This investigation was meant to determine if the range of feed rates utilized in the studies was sufficient

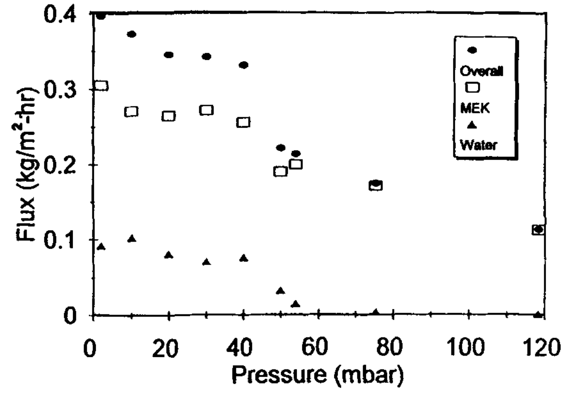

FIG. 7 Flux vs permeate-side pressure using the filled silicone composite membrane for a 2.1% (weight) MEK feed mixture and 50°C.

to insure that there were no significant mass transfer concerns, i.e., concentration polarization, in collecting the previous data. Various researchers (23, 24) have indicated that very dilute feed compositions and low Reynolds numbers contributed to this problem. The concentrations used in this research, the membrane geometry, and the benchmark Reynolds number appear to be satisfactory. Studies were conducted from 1.5 to 9 L/min, and no significant change (<10%) in flux or permeate concentration was observed.

Several types of organophilic membranes were evaluated at benchmark conditions (feed temperature, 50°C; feed concentration, 2.1% MEK; permeate-side pressure, ~2 mbar) to determine their relative performance in terms of flux and selectivity (Fig. 8). Both types of GFT silicone composite membranes were studied; the 1170 silicalite-filled silicone composite membrane as described above and the 1160 which is the standard or unfilled silicone composite membrane. In addition, a poly[1-(trimethylsilyl)-1-propyne] (PTMSP) asymmetric membrane was used in the comparison study.

The 1170 membrane produced a total flux of 0.397 kg/m^2·h as compared to a flux of 1.38 kg/m^2·h for the 1160 membrane, which indicates the 1160 membrane has a flux of approximately 3.5 times that of the 1170 membrane. The permeate concentration obtained for the 1170 membrane was 76.8% compared to 68.4% for the 1160; this yields a selectivity difference of 154 versus 101. These results indicate the difference between the two membranes; the 1170 is a low flux, high selectivity membrane, and the 1160 is a high flux, low selectivity membrane. The relative degrees of

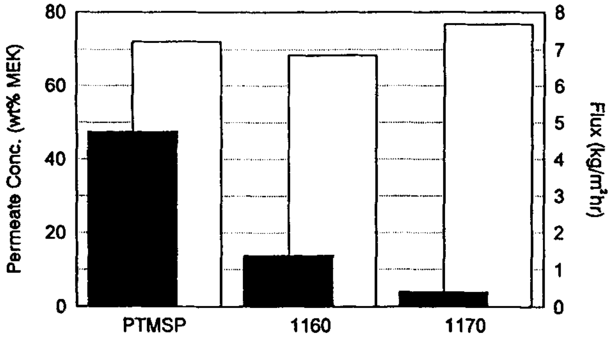

FIG. 8 Permeate MEK concentration (□) and total flux (■) vs organophilic membrane type for a feed mixture of 2.1% MEK, 50°C, and 2 mbar. Membranes: GFT 1170 silicalite filled silicone composite, GFT 1160 unfilled silicone composite, and asymmetric poly[1-(trimethylsilyl)-1-propyne] (PTMSP).

difference depend on the mixture being separated (25). The data indicate that the baseline water flux of the 1170 membrane is 0.092 kg/m^2·h and that the water flux for the 1160 is 0.436 kg/m^2·h. This shows the definite effect the zeolites in the silicone composite structure have on impeding water transport.

Two factors are at work in this comparison study: membrane composite layer thickness and zeolite filling. According to Eq. (1), the thinner the membrane, the higher the flux. Taking the membrane thickness into account, a specific permeation rate can be used which is the product of the component flux and the membrane thickness. The specific water permeation rates are 1.84 and 3.49 kg-μm/m^2·h for the 1170 and 1160 membranes, respectively. These values clearly show how zeolite filling hinders water transport in the 1170 membrane, although zeolite filling at 60% (by weight) does decrease the total membrane flux. Specific MEK permeation rates are 6.10 and 7.55 kg-μm/m^2·h for the 1170 and 1160 membranes, respectively.

The PTMSP membrane produced a much higher flux, 4.75 kg/m^2·h, than the silicone-based membranes. Its selectivity fell between the other two: a permeate MEK composition of 72.2% is produced, which translates to a selectivity of 121. The difference in the polymer is shown here since transport in pervaporation takes place by the solution–diffusion mechanism and therefore solubility and diffusivity in the polymer are different. The specific MEK and water permeation rates for the PTMSP membrane are 6.86 and 2.64 kg-μm/m^2·h, respectively. Although it may appear that the PTMSP membrane is a good choice, it should be noted that the long-term stability of the polymer at high operating temperatures is questionable, and that the membrane utilized was originally developed for gas separation and is not commercially available.

MEK–Water Dehydration Study

Dehydration is an area of much interest in pervaporation because of the potential of direct integration into the numerous process industries that utilize organic solvents in their manufacturing operations. These solvents usually entrain small amounts of water that must be removed if the solvent is to be reused. Additionally, production specifications for the use of organic solvents in industries from pharmaceutical to paints and coatings have stringent requirements for solvent purity.

Dehydration runs were conducted with the GFT 1000 membrane also supplied by Carbone of America. This is considered to be the "standard" GFT membrane for dehydration purposes. It consists of a polyvinyl alcohol (PVA) selective layer on a polyacrylonitrile (PAN) support. The operation of the Zenon bench-scale unit was similar to that for organic permeation runs except for the permeate collection, which had to be performed more frequently since the flux was significantly higher. The same process temperature of 50°C was utilized for the runs to be consistent with the earlier reported organic permeation studies. It should be noted that it is typical industrial practice to operate at a much higher temperature so that flux is enhanced and the design membrane area can be lessened. Experimental studies at various feed concentrations and permeate-side pressures were performed.

Pressure runs were conducted at several permeate-side pressures, and MEK feed concentrations ranged from 91 to 97%. The overall trend observed was that total flux decreased as feed MEK concentration and permeate-side pressure increased. At a permeate-side pressure of 8 mbar and a feed concentration of 95.9% MEK (4.1% water), a total flux of 3.27 kg/m^2·h was obtained (Fig. 9). The permeate MEK concentration was 6.4% (93.6% water), yielding a water selectivity of 340. At 30 mbar and a feed concentration of 90.9% MEK, the permeate concentration was 10.2 and the total flux was 4.4 kg/m^2·h.

MEK–Ethanol Permeation Study

The use of pervaporation technology to effectively separate anhydrous mixtures common to the chemical and petrochemical industry is probably the greatest challenge and holds the most opportunity for long-term commercial growth. The study presented here is an initial investigation into

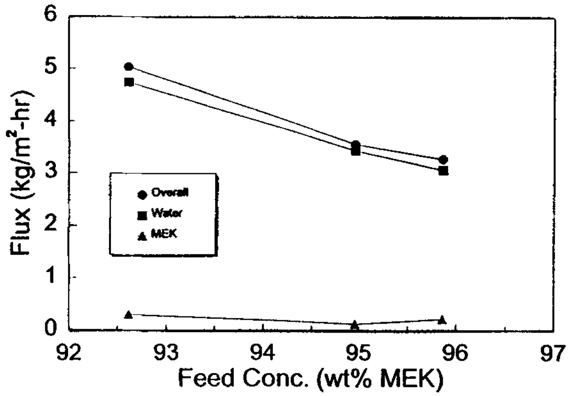

FIG. 9 Flux vs feed MEK concentration using a PVA composite dehydration membrane at a permeate-side pressure of 2 mbar and a feed temperature of 50°C.

this class of pervaporation separations with MEK. Unlike dehydration and organic permeation which use two distinct types of membranes (obviously one selective to water over organics and one selective to organics over water), preferentially permeating a specific organic from an anhydrous mixture requires a more rigorous membrane selection.

As a first step in the direction, the authors used the GFT 1170 silicone composite membrane. The reasoning behind this was that previous studies (13, 26) indicated that in a binary 5% organic–water separation, MEK component flux (kg/m^2·h) is 7.9 times that of ethanol. Although this datum was obtained for a single set of process conditions, it did indicate the possibility of using a standard organophilic membrane which possesses different degrees of permeabilities to various organics. Obviously a membrane engineered by the vendor for this class of separations would give better separation results than the ones reported. Nonetheless, data are presented below to indicate how the relative ability to separate can be demonstrated with a standard organophilic membrane.

The results indicate that the membrane is slightly selective to MEK over ethanol, and the selectivity is much greater when MEK is the minor component (Fig. 10). The ethanol component flux appears to change only slightly (Fig. 11). Runs were conducted at a temperature of 50°C and the lowest permeate-side pressure. At a feed concentration of 24.5% MEK, the permeate was 44.6% and the total flux was 1.65 kg/m^2·h. This represents a selectivity of only 2.5. Using a 50/50 feed mixture, the permeate MEK concentration produced was 66.3% and the total flux was 4.22 kg/m^2·h. Although the organic–water binary separation data may be a good start to determine which component has the greater permeability, they

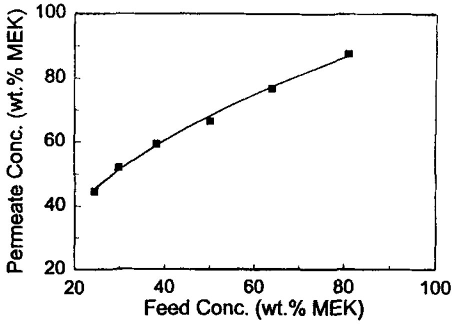

FIG. 10 Permeate MEK concentration vs feed MEK concentration separating an anhydrous mixture of MEK and ethanol. Filled silicone composite membrane operating at 50°C and 2 mbar.

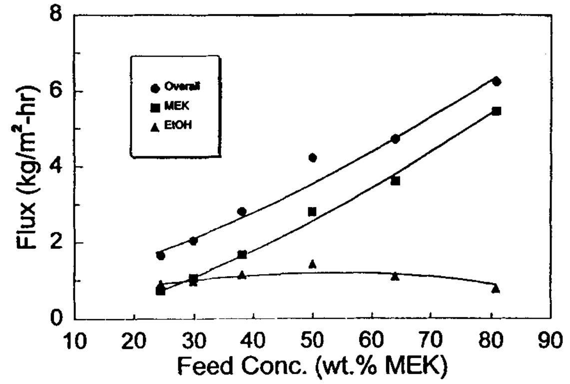

FIG. 11 Flux vs feed MEK concentration separating an anhydrous mixture of MEK and ethanol. Filled silicone composite membrane operating at 50°C and 2 mbar.

appear to be an inadequate predictor of the degree of separation achievable for an anhydrous mixture. Although the membrane was stable for the runs conducted, no long-term stability tests were performed. Future studies are necessary to identify membranes more efficient for this type of separation.

CONCLUSIONS

Pervaporation can be successfully utilized to recover methyl ethyl ketone from various aqueous process mixtures using organic permeation and dehydration membranes. Results with a silicalite-filled silicone composite membrane, GFT 1170, yielded good selectivity and flux for organic permeation from dilute MEK feed mixtures. At a benchmark concentration of 2.1% MEK, 50°C, and permeate-side pressure of 2 mbar, the total flux was 0.397 kg/m^2·h and the permeate concentration was 76.8%. The best processing conditions were obtained at increased temperature and low permeate-side pressure. As feed concentration increased, permeate concentration increased until a plateau was reached around 89%. Feed temperature affected flux in an Arrhenius-type manner with a flux of 1.04 kg/m^2·h obtained at 75°C. As permeate-side pressure increased, flux decreased, and there was a significant effect of pressure on flux as pressure was increased past 40 mbar. Several types of organophilic membranes were compared for this separation. A high flux, low selectivity silicone membrane, GFT 1160, was compared to a low flux, high selectivity membrane, GFT 1170. Both performed their function quite effectively, with the 1160 producing a flux of approximately 3.5 times that of the 1170 with a modest decrease in selectivity. A PTMSP membrane was evaluated, and although it showed promising flux and selectivity, long-term temperature stability was questionable. A polyvinyl alcohol composite membrane, GFT 1000, was employed to successfully dehydrate MEK–water mixtures containing less than 10% water. The separation of an anhydrous mixture of MEK and ethanol was only slightly effective when using a silicone composite membrane. Further membrane development is necessary for this anhydrous separation to optimize the process.

ACKNOWLEDGMENT

The authors gratefully thank Carbone of America for supplying the GFT membranes used in this study.

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Received by editor March 27, 1996
Revision received July 28, 1996