Pervaporative Removal of Organics from Water Using Hydrophobic Membranes. Binary Mixtures

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ABSTRACT

Results of pervaporation experiments are presented for the separation of several polar and nonpolar organic solvents from their aqueous solutions. Three membranes were evaluated: a polydimethylsiloxane (PERVAP-1060) membrane, a PDMS ZSM-5 zeolite filled (PERVAP-1070) membrane, and a poly(ether-*block*-amide) (PEBAX-4033) membrane. The effect of feed composition on flux and selectivity was also investigated. Performance parameters of a given membrane depended both on the kind of the organic solvent and the feed composition. The PERVAP-1070 membrane exhibited the highest selectivity with a separation factor over 900 in contact with a water–butyl acetate mixture. Polar solvents like methanol were also preferentially separated from aqueous solutions, but the separation factors were close to those obtained from liquid–vapor equilibria data. Permeate fluxes of organics increased with increasing feed concentration. Synergetic effects between water and organics fluxes were also observed.

INTRODUCTION

Pervaporation is a membrane separation technique which has become an industrial process in recent years. This process can be used for the dehydration of organic solvents, separation of binary organic–organic mixtures, extraction of organics from aqueous solutions, and/or recovery of aroma compounds in the food and cosmetic industry (1–9).

Removal of organics from aqueous solutions is of particular interest for recycling process water and for the treatment of wastewater. In the environmental industry, removal of volatile organic compounds, like esters, ethers, and/or chlorinated hydrocarbons, by pervaporation is becoming more impor-

TABLE 1
Binary Mixtures and Concentration Ranges Investigated

Binary mixture	Concentration range (wt% organics)
Water–methanol (H$_2$O-MeOH)	0–30
Water–methyl acetate (H$_2$O-MeAc)	0–20
Water–ethyl acetate (H$_2$O-EtAc)	0–5
Water–n-butyl acetate (H$_2$O-BuAc)	0–0.6
Water–methyl t-butyl ether (H$_2$O-MTBE)	0–4

tant and can be more attractive economically than classical methods (6–10). To optimize the overall separation process, pervaporation is usually combined with other separation methods (11).

The objective of this work was to investigate the performance parameters of different organophilic membranes in contact with several binary water–organic mixtures, i.e., water–methanol, water–methyl acetate, water–ethyl acetate, water–n-butyl acetate, and water–methyl t-butyl ether. Organic components of these binary mixtures can be found in effluents from the pharmaceutical, textile, and/or petrochemical industries.

EXPERIMENTAL

The concentration ranges investigated for each binary mixture are summarized in Table 1. Table 2 presents chosen physicochemical properties of water and the organic solvents investigated.

TABLE 2
Chosen Physicochemical Data of Investigated Solvents (12, 13)

Solvent	Molecular weight (g/mol)	Molar volume (cm^3/mol)	Solubility in water (g/100 g H$_2$O)	Dielectric constant ε (–)	Solubility parameter δ [(cal/cm^3)$^{0.5}$]
Water	18	18.0	—	81.0	23.4
MeOH	32	40.5	∞	31.2	14.3
MeAc	74	92.5	31.9	7.1	9.6
EtAc	88	98.0	7.9	6.1	9.1
BuAc	116	132.0	0.7	5.0	8.5
MTBE	88	119.0	5.1	$\approx$3	—

TABLE 3
Characteristics of Pervaporation Membranes Investigated

Membrane	Skin layer	Producer
PERVAP-1060	PDMS,[a] $d = 8$ μm	Sulzer Chemtech Membrantechnik AG (Germany)
PERVAP-1070	PDMS filled with ZSM-5[b] zeolite, $d = 10$ μm	Sulzer Chemtech Membrantechnik AG (Germany)
PEBAX-4033	PEBA,[c] $d = 80$ μm	GKSS (Germany)

[a] PDMS, polydimethylsiloxane:

$$\left[\begin{array}{c} CH_3 \\ | \\ -Si-O- \\ | \\ CH_3 \end{array} \right]_n$$

[b] ZSM-5, zeolite: $Na_n[Al_nSi_{(96-n)}]O_{192}] \sim 16H_2O$, $n < 27$

[c] PEBA, poly(ether-*block*-amide):

$$HO- \left[\begin{array}{ccccccc} C & -PA- & C & -O- & PE & -O \\ \| & & \| & & & \\ O & & O & & & \end{array} \right]_n -H$$
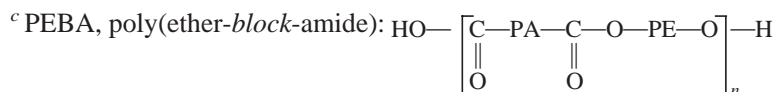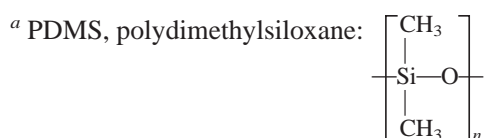
Membranes

Three types of hydrophobic membranes were used in this study: PERVAP-1060 (Sulzer, Germany), PERVAP-1070 (Sulzer, Germany), and PEBA-4033 (GKSS, Germany). All these membranes are composite. The PERVAP-1060 membrane possesses a selective layer made of polydimethylsiloxane (PDMS). The PERVAP-1070 membrane is composed of a thin PDMS layer with hydrophobic ZSM-5 zeolite particles incorporated into it to increase selectivity. The selective top layer of PEBAX-4033 membrane is made of polyether–polyamide block copolymer (PEBA). PEBA is a thermoplastic elastomer which consists of rigid polyamide linear blocks and flexible polyether linear blocks (14, 15). The characteristics of the investigated membranes are summarized in Table 3.

Pervaporation Measurements

Pervaporation experiments were carried out in the laboratory-scale pervaporation system presented in Fig. 1. The system included a temperature-controlled feed vessel. Feed solution circulated over the membrane. The permeate was collected in cold traps cooled by liquid nitrogen. During experiments the upstream pressure was maintained at atmospheric pressure while the downstream pressure was kept below 1 mbar by using a vacuum pump. The
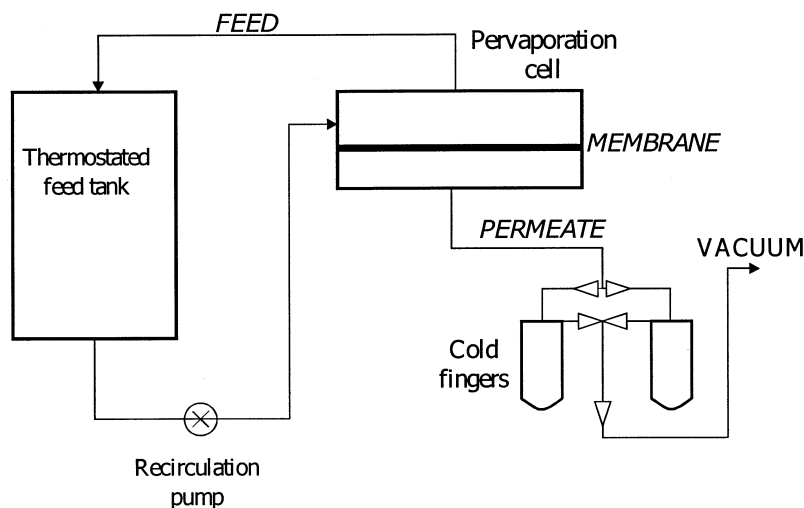
FIG. 1   Schema of the laboratory-scale pervaporation setup.

permeate flux was determined by weight, whereas the feed and permeate compositions were determined by using a gas chromatograph (Varian 3300).

The performance properties of a given pervaporation membrane are defined by the separation factor α (Eq. 1) and permeate fluxes J (16).

$$\alpha_{\text{org/water}} = \frac{(c_{\text{org}}/c_{\text{water}})''}{(c_{\text{org}}/c_{\text{water}})'} \tag{1}$$

The effect of feed composition on flux and selectivity was determined for each membrane while it was in contact with different binary organics–water mixtures.

RESULTS AND DISCUSSION

Permeability of PDMS Membranes toward Pure Solvents

Table 4 presents the permeability properties of both PERVAP membranes in contact with pure water, methanol, and methyl acetate. Permeability coefficient P was calculated according to

$$J = P\frac{\Delta p}{d} \tag{2}$$

where J = permeate flux
P = permeability coefficient
Δp = vapor pressure difference
d = thickness of the selective layer

TABLE 4
Pervaporation Permeability of PERVAP-1060 and PERVAP-1070 Membranes in Contact with Pure Solvents

	PERVAP-1060		PERVAP-1070	
Solvent	J $[10^{-3}\ mol\cdot m^2\cdot s^{-1}]$	P $[10^{-1}\ mol\cdot N^{-1}\cdot s^{-1}]$	J $[10^{-3}\ mol\cdot m^2\cdot s^{-1}]$	P $[10^{-1}\ mol\cdot N^{-1}\cdot s^{-1}]$
H_2O	2.42	0.19	0.82	0.06
MeOH	8.05	0.13	3.66	0.06
MeAc	14.74	0.12	12.92	0.11

Comparing PERVAP-1060 and PERVAP-1070 membranes, it is seen that permeate fluxes through the latter are smaller regardless of the kind of solvent. This is the effect of zeolite filling. However, when a nonpolar solvent like MeAc contacts the PERVAP-1070 membrane, the permeability is only slightly lowered. The different behaviors of the PERVAP-1070 membrane in contact with polar (water, MeOH) and nonpolar (MeAc) solvents can be explained by the enhanced sorption of MeAc into the zeolite pores and by the facilitated transport of sorbed MeAc through both zeolite pores and polymer. When a polar solvent is transported, its molecules are not sorbed nor transported through the zeolite pores. The diffusion path of water and methanol molecules through a zeolite-filled membrane is more tortuous than through an unfilled membrane (Table 4). These results suggest also that the ZSM-5 filling of the PERVAP-1070 membrane should enhance the selectivity of this membrane in contact with water–organic mixtures compared to the PERVAP-1060 membrane if the organic component possesses low polarity and its dimensions are smaller than the diameter of the zeolite pores (17).

Separation of Water–Methanol Mixtures

Separation of water–alcohol mixtures by pervaporation has been investigated by many workers, with special attention paid to the water–ethanol system. However, only a few works have been devoted to the water–methanol mixture (18–22). Methanol often occurs as one of the components in multicomponents mixtures in waste and process waters.

The separation characteristics of all the investigated membranes in contact with water–methanol mixtures are presented in Fig. 2. It is seen that the concentration of methanol in the permeate is always higher than in the feed, i.e., methanol permeates preferentially through these membranes. Moreover, comparing the pervaporation separation with liquid–vapor equilibrium, one can state that the selectivity of the PERVAP-1060 membrane is practically equal to the distillation selectivity. The selectivity factor α is in the 6–7 range for the
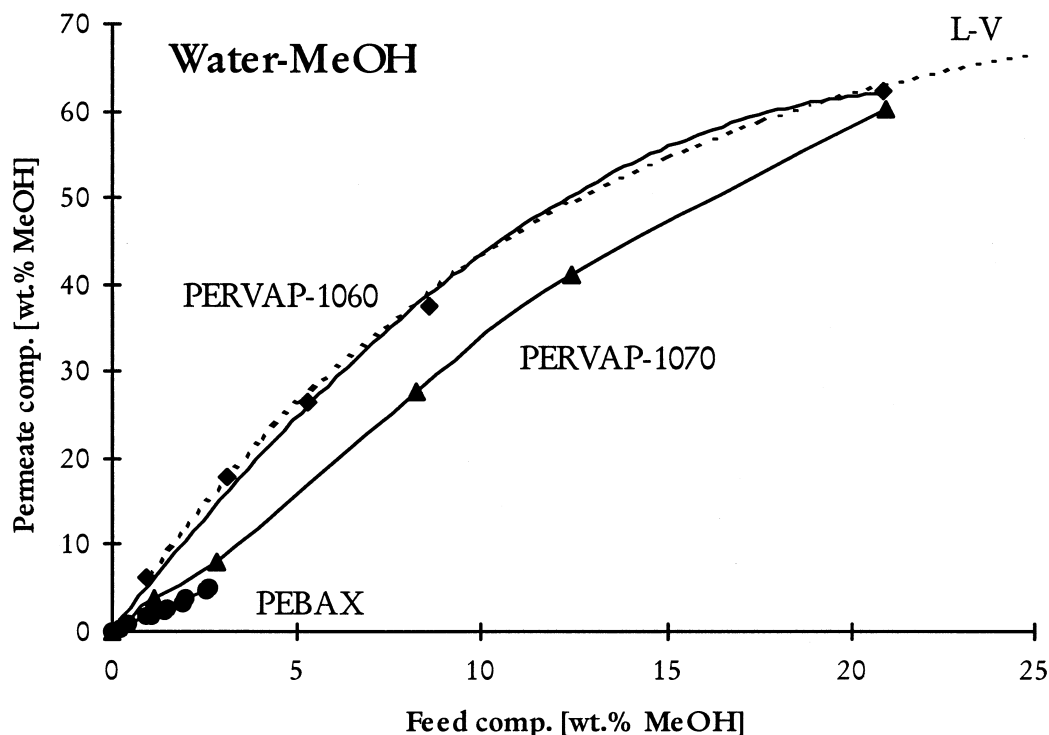
FIG. 2   McCabe–Thiele selectivity diagram for investigated membranes in contact with water–MeOH mixture.

PERVAP-1060 membrane, 3–5 for the PERVAP-1070 membrane, and 2 for the PEBAX-4033 membrane. The lower selectivity of the PERVAP-1070 membrane compared to the PERVAP-1060 membrane can be explained by the fact that methanol molecules have too high a polarity to pass through the highly hydrophobic zeolite particles (17). The zeolite pores are impenetrable to both water and methanol molecules during pervaporation. Moreover, the tortuosity introduced by zeolite particles obstructs methanol transport more than water transport. As a consequence, selectivity of the PERVAP-1070 membrane is lowered compared to the PERVAP-1060 membrane, but the former still preferentially separates methanol from water ($\alpha > 1$). These results are very consistent with the results obtained for pervaporation of pure solvents (Table 4).

Figure 3 presents the concentration dependence of the permeate fluxes for the PERVAP-1060 membrane in contact with water–methanol mixtures at 303 K. The methanol flux increases practically linearly with its concentration in the feed whereas the flux of water slightly decreases in the investigated concentration range. The concentration dependence of the permeate fluxes through the PERVAP-1070 membrane at 303 K is shown in Fig. 4. The permeate fluxes of the zeolite-filled PERVAP-1070 membrane are lower than those of the unfilled
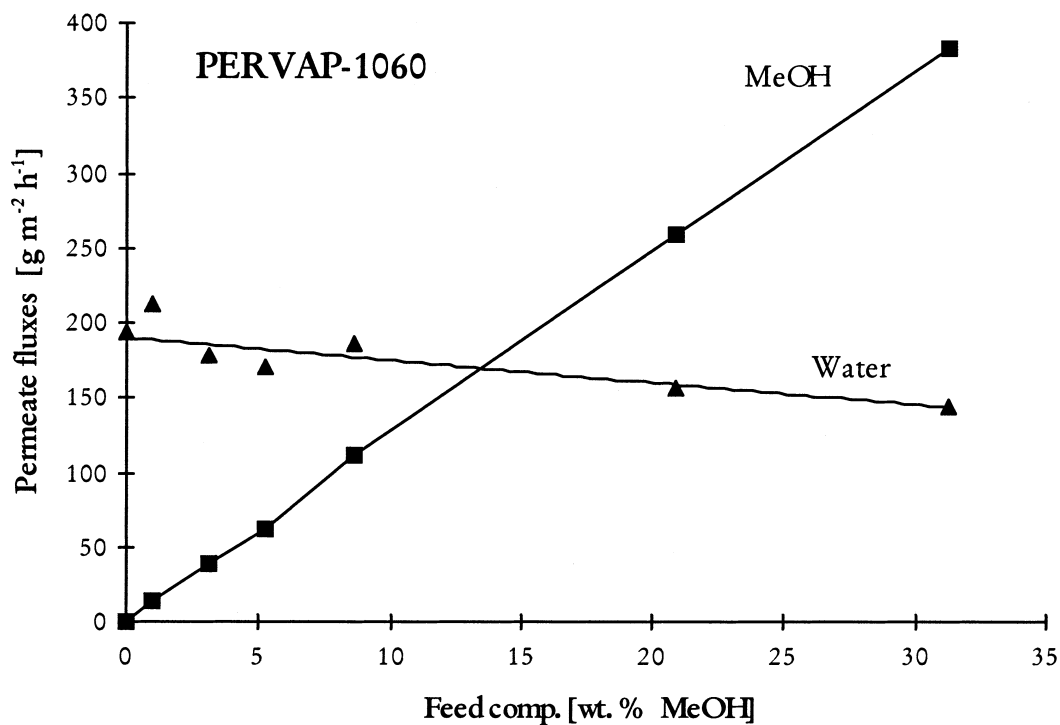
FIG. 3   Pervaporation permeate fluxes of water and MeOH in contact with PERVAP-1060 membrane.
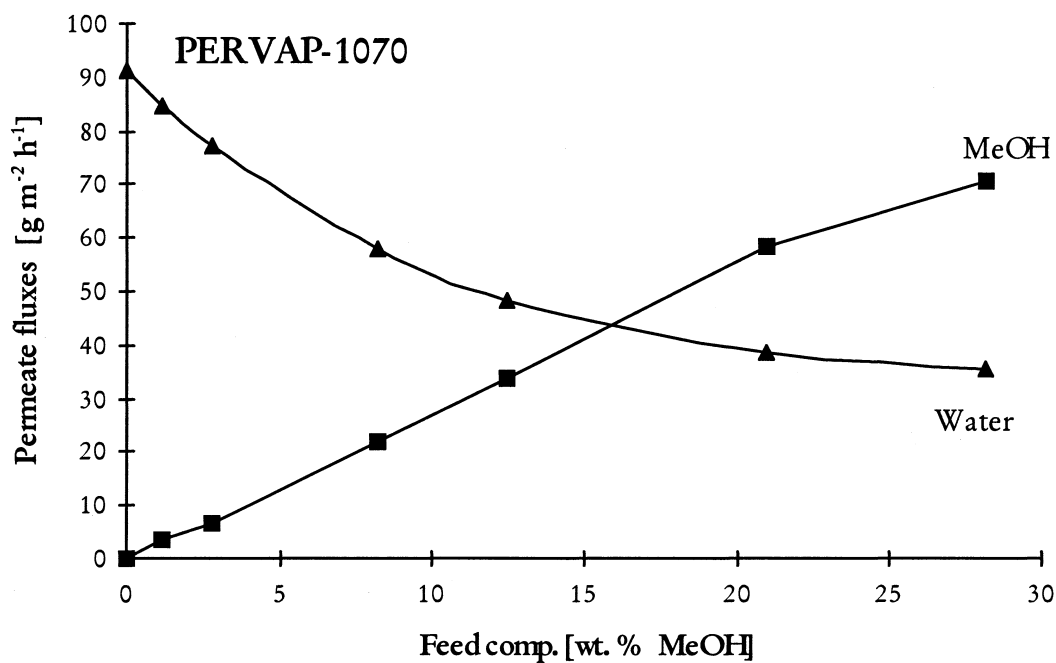

FIG. 4   Pervaporation permeate fluxes of water and MeOH in contact with PERVAP-1070 membrane.
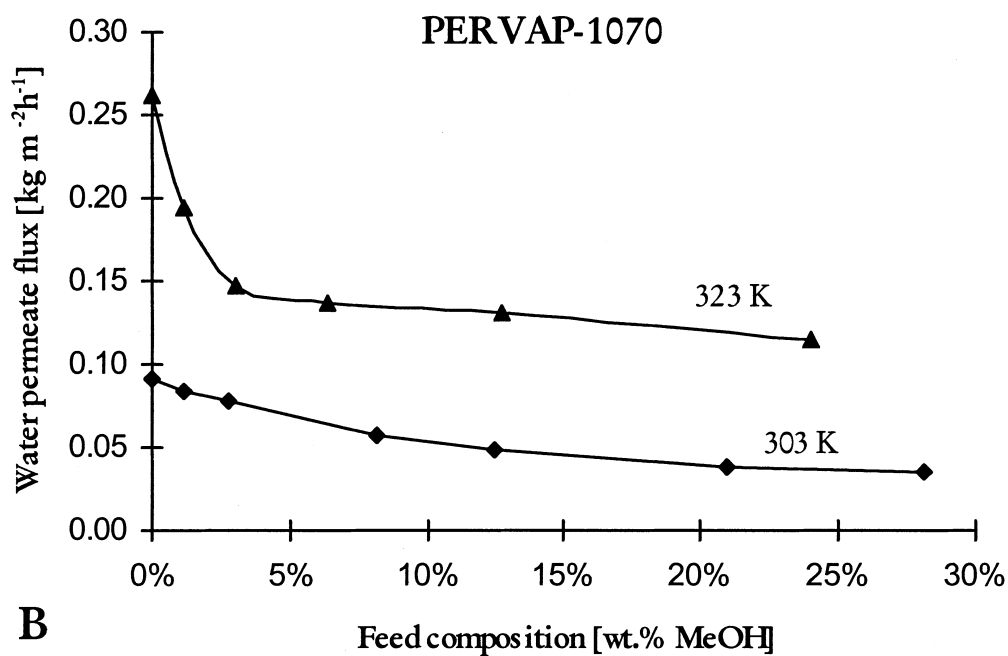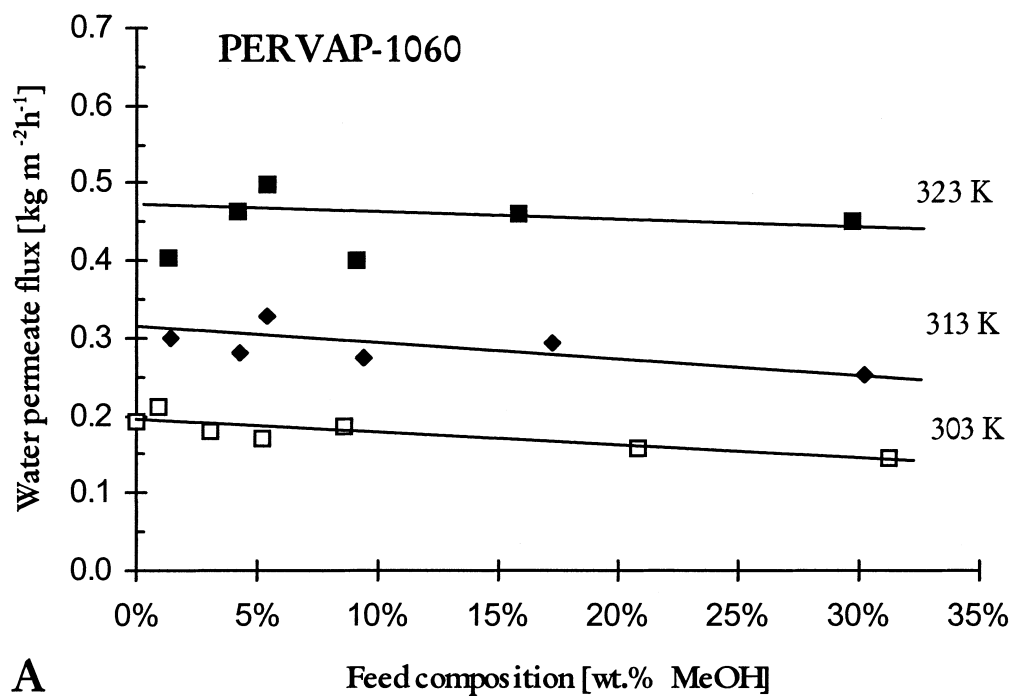
FIG. 5   PV permeate fluxes of water vs feed composition at different temperatures. A: Water–MeOH mixture in contact with PERVAP-1060 membrane. B: Water–MeOH mixture in contact with PERVAP-1070 membrane.

PERVAP-1060 membrane (Fig. 5). This is caused by the fact that zeolite particles present in the 1070 membrane obstruct transport due to the tortuosity introduced. Moreover, the active layer in the zeolite-filled membranes is usually thicker than that in the unfilled ones, causing also the reduction of flux (Table 3). The permeate flux of methanol through both the PERVAP-1060 and PERVAP-1070 membranes increases linearly with increasing feed concentration. On the other hand, the concentration dependence of water flux is different for these two membranes. Water flux through the PERVAP-1060 membrane slightly decreases over the concentration range investigated (Fig. 5A) whereas water flux through the PERVAP-1070 membrane descends suddenly up to a concentration of 0.5–2 wt% MeOH in the feed (Fig. 5B). This relation is much better pronounced at higher temperatures. The permeate fluxes increase with increasing temperature, but the general relations between them remain the same (Fig. 5). Calculated activation energies for the transport of water and methanol molecules through the PERVAP-1060 membrane are 43 and 33 kJ/mol, respectively. This indicates that with increasing temperature the selectivity of the PERVAP-1060 membrane will slightly decrease.

Permeate fluxes through the PEBAX-4033 membrane are presented in Fig. 6. Although the flux of MeOH through this membrane is much lower than the flux of water, PEBAX-4033 still transports methanol molecules preferentially
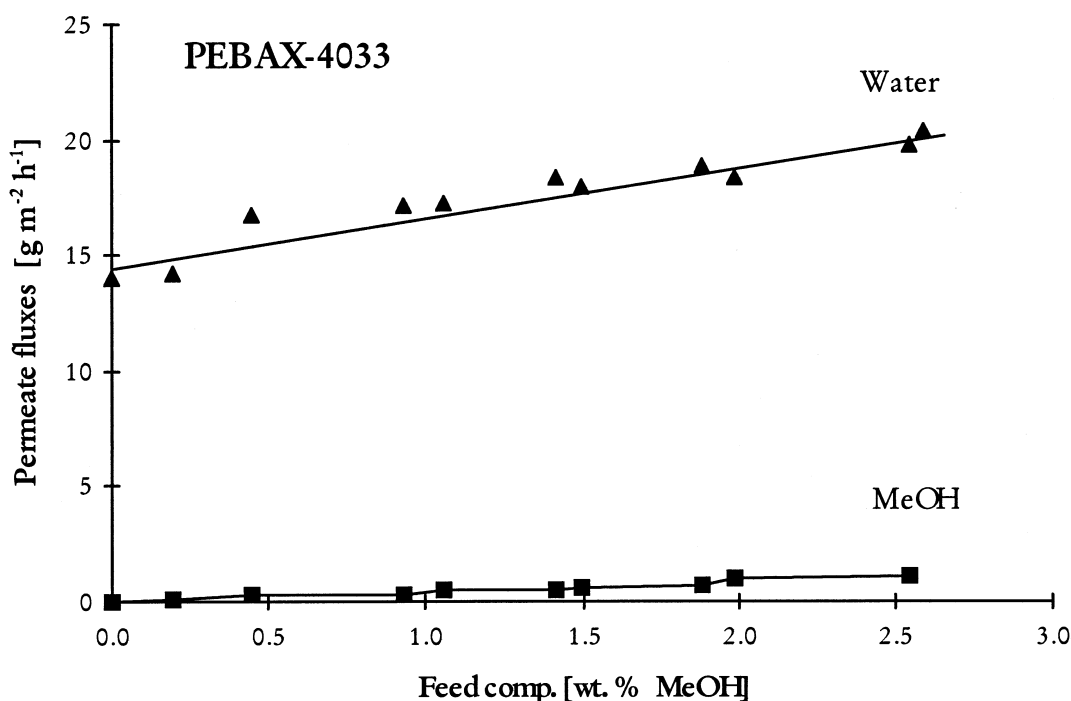

FIG. 6   Pervaporation permeate fluxes of water and MeOH in contact with PEBAX-4033 membrane.

(Table 4 and Fig. 2). By comparing these transport results with the results of Suzuki et al. (19), we find that the concentration relation of water and methanol fluxes through the PEBAX-4033 membrane resembles the relations found for the water–MeOH system and the poly(γ-methyl L-glutamate)/PMLG membrane (19).

Separation of Water–Esters Mixtures

Separation properties of the investigated membranes in contact with water–esters mixtures are presented in Fig. 7. The selectivity of a given membrane decreases with increasing polarity of an ester. However, for the PERVAP-1060 and PERVAP-1070 membranes (Fig. 7A, B), differences in selectivities between esters are less pronounced than for the PEBAX-4033 membranes (Fig. 7C). By comparing the selectivity of the investigated membranes toward a given ester it can be seen that the zeolite filling of PERVAP-1070 enhances separation and this membrane is the most selective one (Fig. 8). The PEBAX-4033 membrane is less selective than the PERVAP-1060 membrane in contact with water–MeAc and water–EtAc mixtures (Fig. 8A, B) and becomes more selective than the latter membrane in contact with water–BuAc mixtures (Fig. 8C). The strong influence of the solvent polarity on PEBAX-4033 properties can result from the chemical structure of this membrane in which more polar polyamide blocks are mixed with less polar polyether blocks.

The transport properties of investigated membranes are presented in Figs. 9–11. The permeate flux of an organic component increases as its content in the feed mixture increases. Such an effect of the organic feed concentration on the pervaporation of aqueous mixtures through organophilic membranes was also found in other systems (18–24).

For a given membrane the pervaporation flux of esters increases with decreasing polarity of an ester, and this relation is much more pronounced for the PEBAX-4033 and PERVAP-1070 membranes than for the siloxane PERVAP-1060 one. The permeation rates and α separation factors for aqueous esters solutions through organophilic membranes increase with the molecular size of the ester. These permeation and separation relations suggest that both the solubility and the diffusivity of the permeating molecules through the membrane are significant. By comparing the values of the solubility parameter (δ) of the esters (Table 2) with the value of the solubility parameter of PDMS [$\delta_{PDMS} = 8.1$ (cal/cm^3)$^{0.5}$], it is seen that PDMS PERVAP-1060 has the strongest affinity for BuAc. Moreover the degree of swelling of BuAc in this membrane exceeds that of MeAc and EtAc. High swelling in aqueous BuAc mixtures also causes the PDMS chains to become more flexible, and so the transport of BuAc molecules is faster than the transport of MeAc and EtAc molecules. Consequently, BuAc can be permeated through the PERVAP-1060 membrane much easier, and the highest selectivity for BuAc occurs.
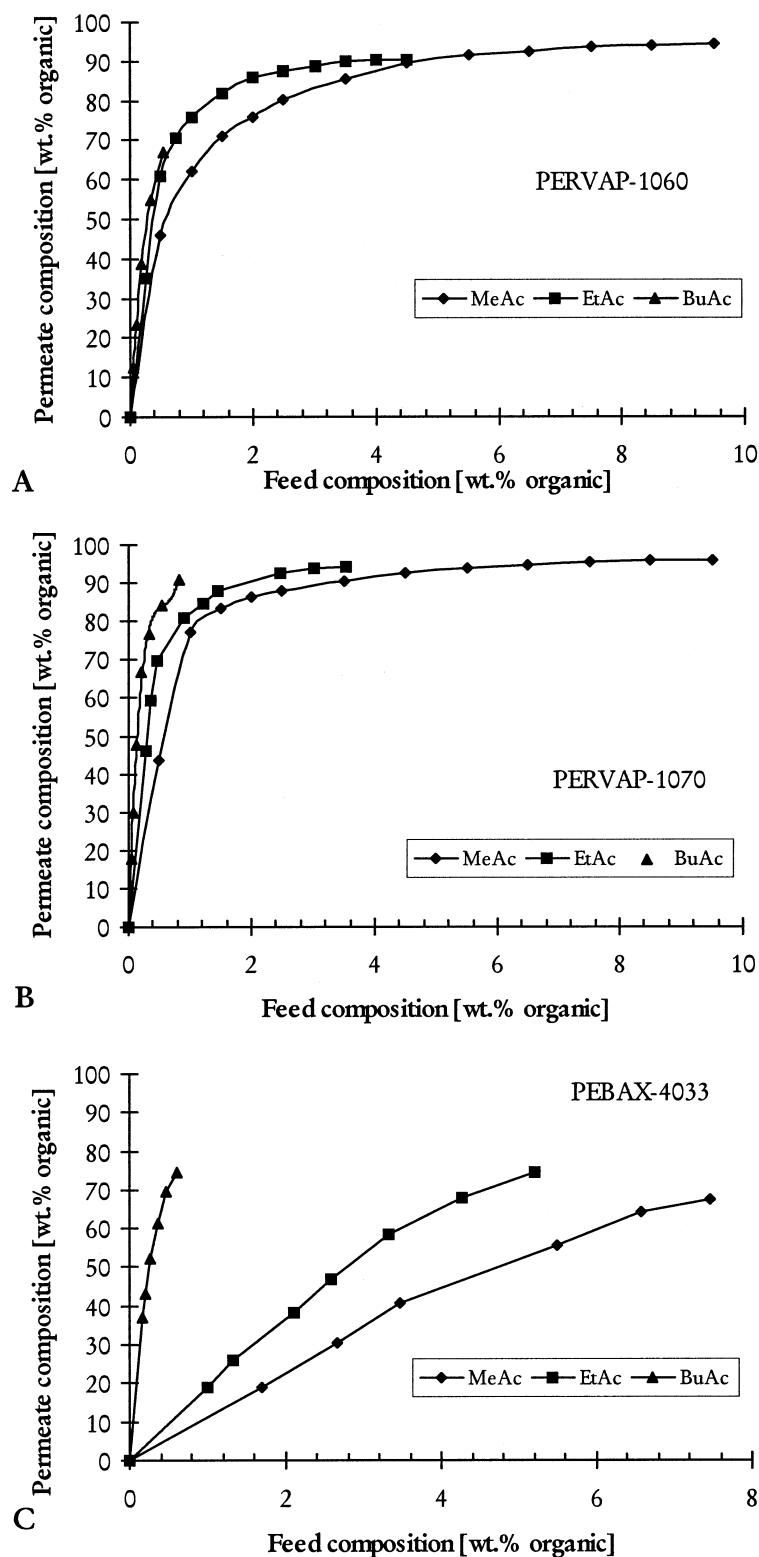
FIG. 7   McCabe–Thiele selectivity diagram for investigated membranes in contact with water–ester mixtures. A: PERVAP-1060 membrane. B: PERVAP-1070 membrane. C: PEBAX-4033 membrane.
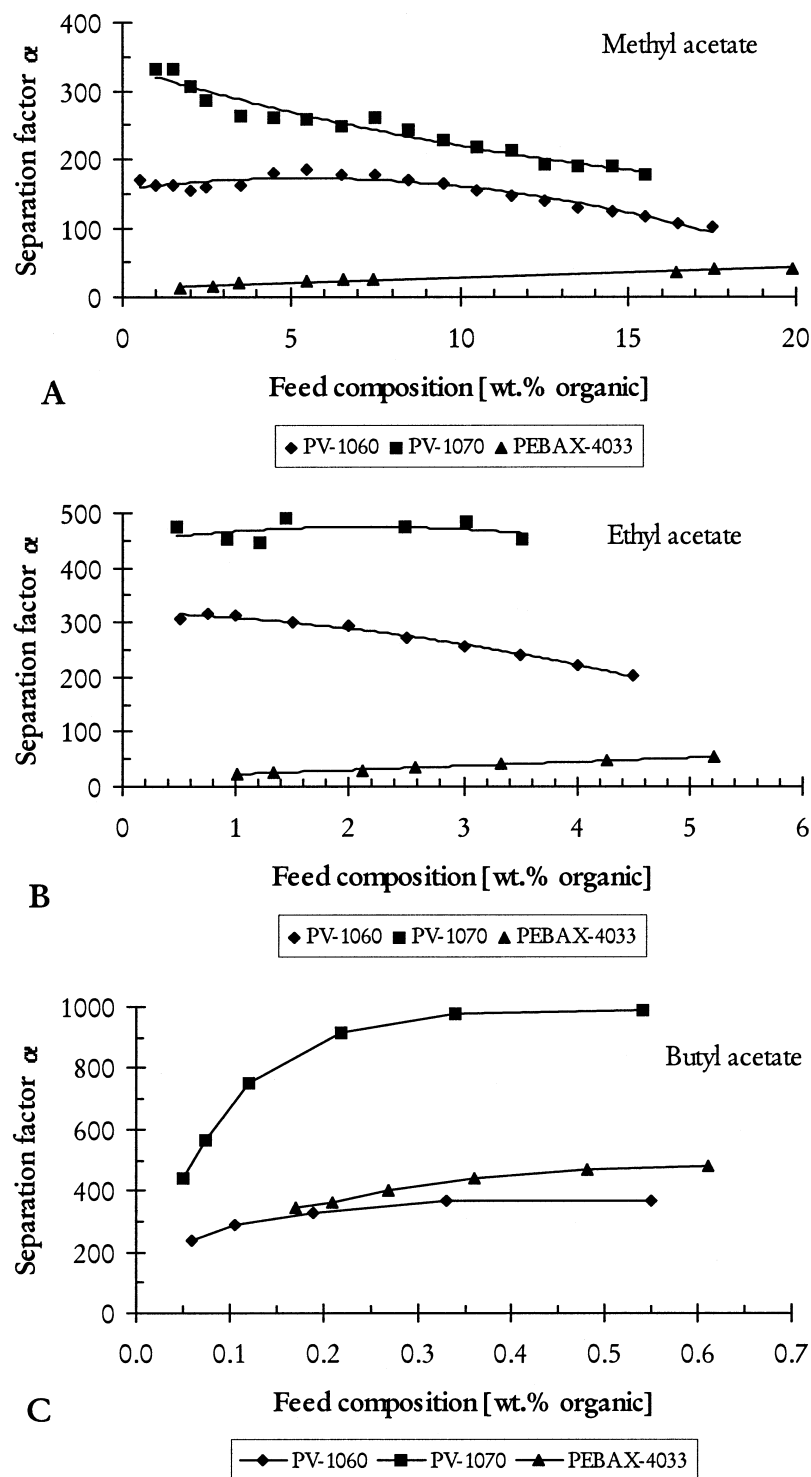
FIG. 8   Concentration dependence of separation factor for water–ester mixtures in contact with a given membrane. A: PERVAP-1060 membrane. B: PERVAP-1070 membrane. C: PEBAX-4033 membrane.
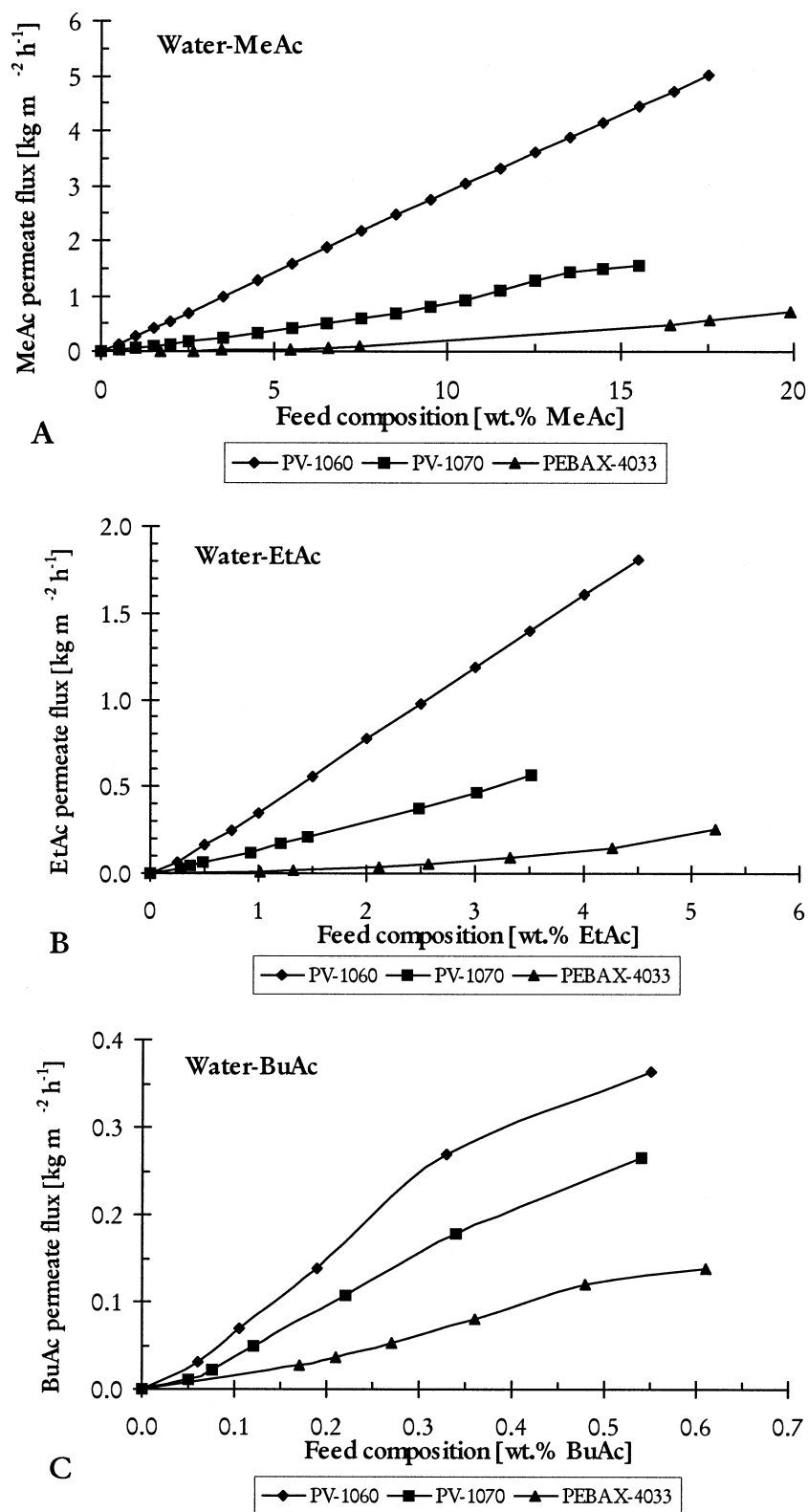
FIG. 9   Pervaporation permeate fluxes of esters vs feed composition in contact with investigated membranes. A: Water–MeAc mixture. B: Water–EtAc mixture. C: Water–BuAc mixture.

For a given water–ester mixture the organic permeate flux is highest for the PERVAP-1060 membrane and lowest for the PEBAX-4033 membrane. Similar relations were found for the water–methanol mixture. However, the thickness of the selective top layer is different for each membrane (Table 3). To compare ester permeate fluxes regardless of the differences of the top layer, the normalized fluxes (i.e., fluxes of the permeate through a membrane with a hypothetical thickness of 1 μm) of BuAc through the investigated membranes are presented in Fig. 10. It is seen that the normalized permeate flux of BuAc molecules through PEBAX-4033 is the highest, and that permeate normalized fluxes of BuAc through PDMS (PERVAP-1060) and zeolite-filled PDMS (PERVAP-1070) membranes are close to each other. Consequently, zeolite filling of the PERVAP-1070 membrane does not hinder the transport of BuAc molecules but does retard the transport of water molecules (the separation factor of BuAc is higher for the PERVAP-1070 membrane compared to the PERVAP-1060 membrane—Fig. 8).

The affinity of water to the hydrophobic membranes is very low, and consequently the flux of water molecules through such membranes should be low and proportional to the feed composition. The presence of the organic component in the feed mixture and in the membrane will affect both the sorption and the water permeate flux. Figure 11 presents the concentration dependencies of water permeate fluxes through the investigated membranes. It is seen that in the
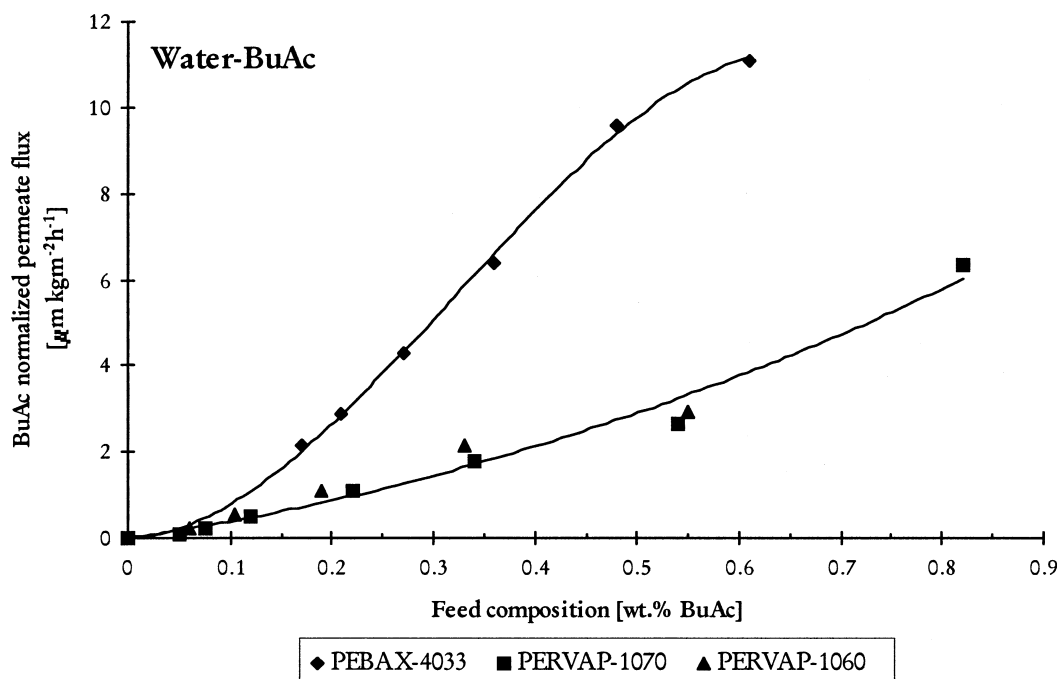

FIG. 10   Normalized permeate fluxes of BuAc through investigated membranes vs feed composition.
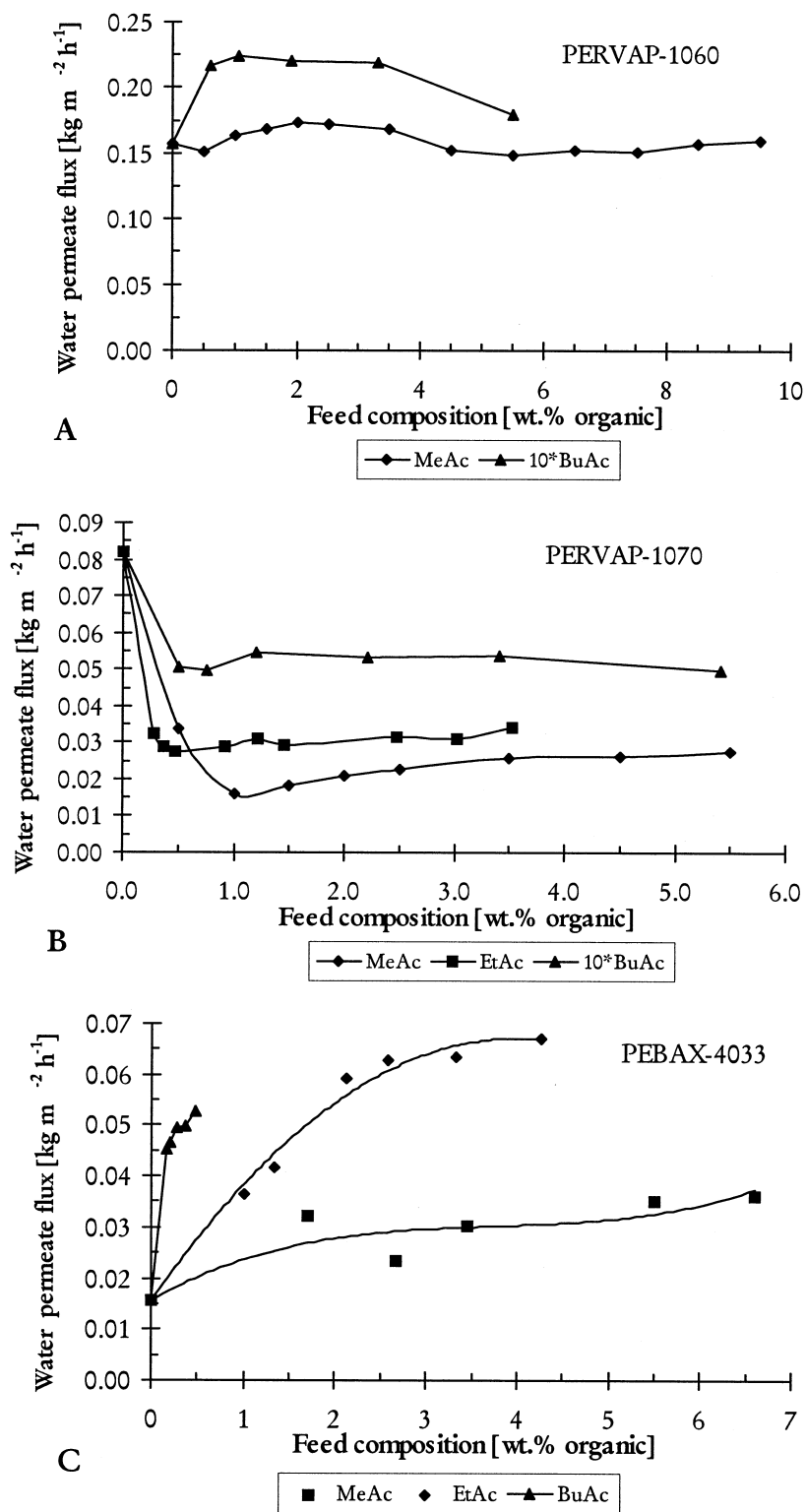
FIG. 11   Pervaporation permeate fluxes of water vs feed composition in contact with investigated membranes. A: PERVAP-1060 (note: the feed concentration of BuAc was multiply by 10). B: PERVAP-1070 (note: the feed concentration of BuAc was multiply by 10). C: PEBAX-4033.

case of unfilled membranes (PERVAP-1060 and PEBAX-4033) the flux of water increases with the increasing organic content in the feed. Moreover, the flux of water is affected by the presence of a given ester to a larger extent as the polarity of the ester is lower (Fig. 11A, C). The results presented in Fig. 11(B) show the influence of both the zeolite filling in the PERVAP-1070 membrane and the ester polarity on the water transport. In this case, water flux passes through a minimum and the feed concentration at which this minimum occurs depends of the kind of ester: 1 wt% for the water–MeAc mixture, 0.4 wt% for the water–EtAc mixture, and 0.07 wt% for the water–BuAc mixture. Water flux is suppressed most strongly when a membrane contacts the water–MeAc mixture (Fig. 11B). All these results suggest the strong synergetic effects occurring during transport of ester and water molecules through a membrane.

Separation of Water–MTBE Mixtures

MTBE is produced on a large scale from isobutylene and methanol and is used as a fuel enhancer. A number of papers have been published on the application of pervaporation to enhance the synthesis of MTBE by the separation of the methanol–MTBE mixture (25–30). However, MTBE, which was appreciated for its proecological properties as a lead alkyls replacer, can also cause severe air and water pollution and is suspected of being a carcinogenic chemical (31, 32). Pervaporation can be a method used to diminish water pollution by MTBE. Pervaporation results of MTBE removal from water are presented in Figs. 12–14. Similar to other investigated systems, all organophilic
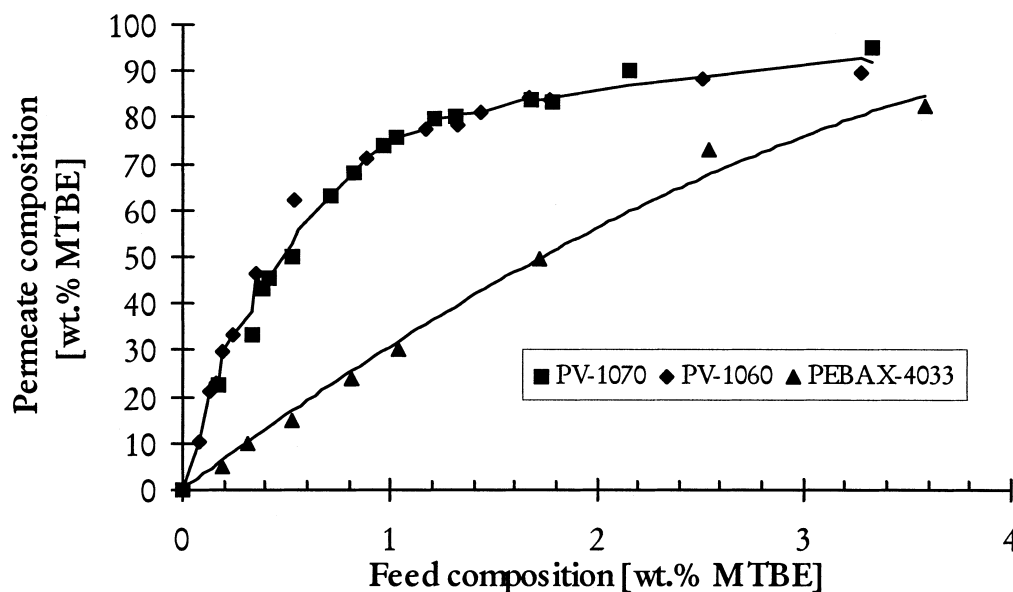

FIG. 12   McCabe–Thiele separation diagram of investigated membranes in contact with water–MTBE mixtures.
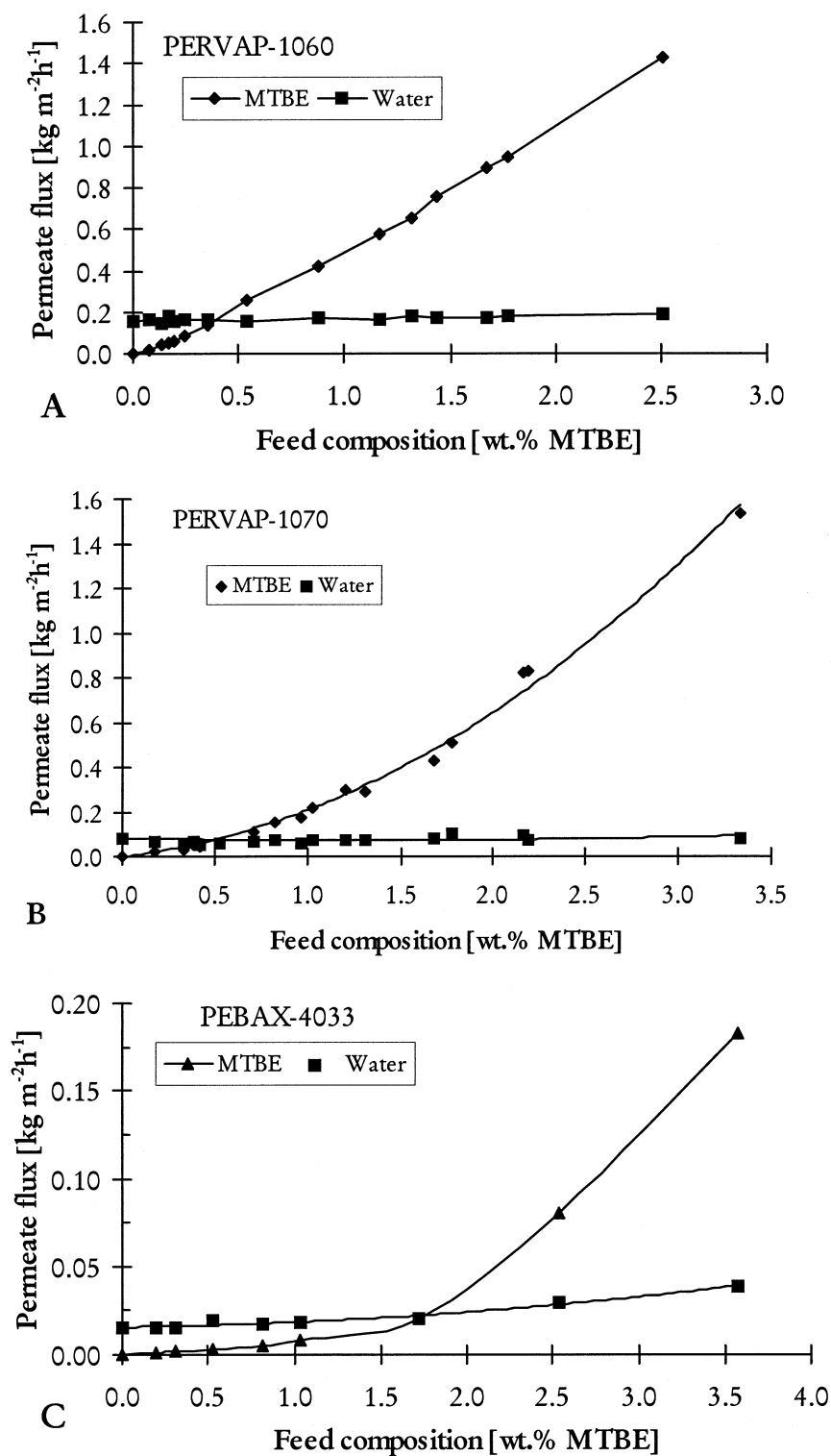
FIG. 13 Pervaporation permeate fluxes of water and MTBE through investigated membranes vs feed composition. A: PERVAP-1060 membrane. B: PERVAP-1070 membrane. C: PEBAX-4033 membrane.
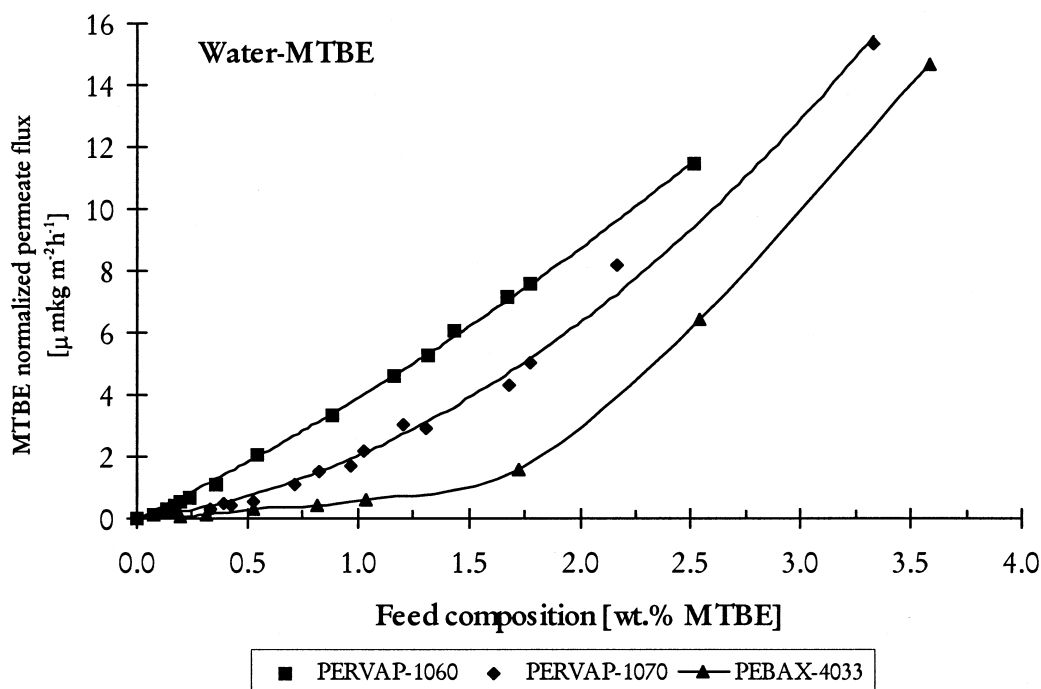
FIG. 14   Normalized permeate fluxes of MTBE through investigated membranes vs feed composition.

membranes selectively separate MTBE from water (Fig. 12). It is worth noting that the separation properties of PERVAP-1060 and PERVAP-1070 membranes are practically identical. This suggests that zeolite filling of PERVAP-1070 does not enhance the separation of MTBE from water. The reason may be either the branched shape and relatively high volume of MTBE molecules (Table 1) or differences in the polarity between zeolite particles and MTBE molecules.

The transport results obtained for aqueous MTBE solutions are similar to those obtained for other water–organic systems investigated (Fig. 13). The permeation fluxes of MTBE through all the investigated membranes increase as the MTBE concentration in the feed is increased. The normalized permeate fluxes of MTBE through the investigated membranes are presented in Fig. 14. The MTBE molecules permeate faster through the PERVAP-1060 membrane than through the PERVAP-1070 membrane. This confirm the suggestion that the zeolite filling of PERVAP-1070 increases the transport path of MTBE molecules and does not influence the selectivity (Fig. 12). The concentration dependence of the water permeation flux resembles that obtained for water–methanol and water–esters mixtures, indicating couplings between the fluxes of water and MTBE.

CONCLUSIONS

Pervaporation is a type of membrane separation which can be applied in the removal of volatile organic compounds from water. The selective separation of organics from aqueous mixtures has been studied using three types of organophilic membranes. All the investigated membranes were selective in the removal of VOCs from water. The zeolite-filled PERVAP-1070 membrane was found to be the most effective in terms of selectivity. The PERVAP-1060 membrane also showed very good selective and transport properties. The PEBAX-4033 membrane was found to be highly selective in contact with the water–BuAc mixture.

It was also found that pervaporation can be regarded as an effective method in the removal of MTBE from contaminated groundwater. In this application another separation technique should probably be coupled with pervaporation to attain a satisfactory level of purification.

Water flux through hydrophobic membranes was greatly affected by the kind and amount of organic component in the binary mixture.

ACKNOWLEDGMENTS

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