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Pervaporation of Aroma Compounds Using Virgin and Silicalite-Filled Organophilic Membranes: Effect of Aroma Compound Structure and Comparison with Distillation Selectivity

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ABSTRACT

Pervaporative recovery of individual aroma compounds having different functional groups is studied using polydimethylsiloxane (PDMS) as well as silicalite filled PDMS (PDMS SA-5) membranes. The aroma compounds studied are linalool, citronellal, menthone, and β-ionone. The performance of both the membranes was compared. The effective selectivity of organophilic membranes is compared with the vapor–liquid equilibria (VLE) selectivity estimated using thermodynamic data. Effect of the nature of each aroma compound on its pervaporation performance

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is also discussed. Sorption effect was found to be predominant in determining the pervaporation selectivity.

Key Words: Organophilic pervaporation; Aroma compounds; Vapor–liquid equilibrium; PDMS membrane.

INTRODUCTION

Pervaporation is a membrane separation process in which a liquid feed mixture is separated by partly vaporizing it through a nonporous permselective membrane. The driving force is the chemical potential gradient, which is created by applying vacuum by continuous pumping at the permeate side of the membrane.[1] This membrane process is different from the other membrane processes since there is phase change at the downstream side of the membrane.

Pervaporation is widely used for recovery of volatile organic compounds due to its high separation efficiency, mild operating conditions, and simple modular nature.

Pervaporation is an ideal separation process due to its following characteristics:

1. In the case of azeotropic mixture (e.g., water–ethanol) pervaporation can be used without cosolvent unlike azeotropic distillation.
2. Recovery of aroma compounds without degradation of temperature sensitive compounds as pervaporation is generally applied at ambient temperature.
3. The coupling of pervaporation with other processes gives high separation efficiency.

In the production of essential oils many oxygenated organic compounds are lost in steam condensate.[2] These oxygenated compounds (alcohols, aldehydes, ketones, esters, etc.) are mainly responsible for the natural aroma of essential oils.[3] For recovery of these aroma compounds, hydrophobic membranes such as polydimethylsiloxane (PDMS) and silicalite filled PDMS (PDMS SA-5) have been used.[2] Many essential oils produced by steam distillation from botanical sources are widely used in the perfume and flavor industries. More recently, pervaporation has been applied to the extraction of the aroma compounds either produced from biotechnology, which are highly valuable because of their natural origin[4] or recovered from perfumery wastes. The pervaporative recovery of ethanol has also been studied.[5]

Pervaporation finds another application in removing traces of organics from aqueous solution using unfilled hydrophobic membranes[6] as well as adsorbent-filled membranes.[7] Many fermentation–pervaporation couplings have been applied for the simultaneous production–extraction of organic compounds such as ethanol,[8] butanol,[9] acetone,[9] acetoin, and butanediol.[10] The influence of the nature of the aroma compounds and other compounds present in the liquid feed, the nature of pervaporation membranes, as well as the importance of engineering aspects of pervaporation process have been discussed.[11]

The present study deals with recovery of aroma compounds like linalool, citronellal, menthone, and β-ionone from aqueous solution using PDMS and silicalite (SA-5) filled PDMS membranes. The properties of all the compounds studied are given in Table 1. The pervaporation performance of both the membranes is compared for each aroma compound. Further, the pervaporation selectivity of both the membranes has been compared with vapor–liquid equilibria (VLE) selectivity.

THEORY

The transport of the permeate through a dense membrane is usually described by the solution-diffusion model.[12] The following assumptions are made in this model.

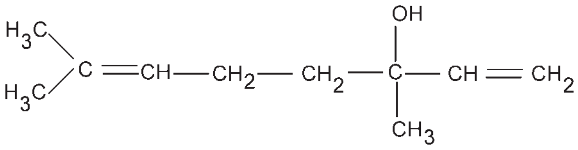
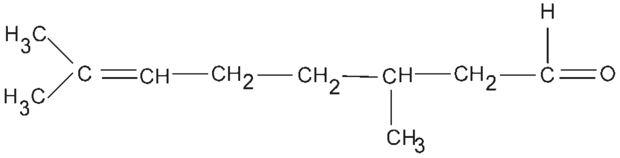
1. Preferential sorption of the solute component occurs at the feed side of the membrane, giving rise to sorption selectivity.
2. Diffusion of the solute component across the membrane.
3. Desorption of the solute component on the downward face of the membrane.

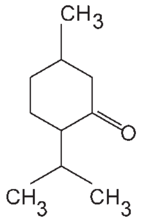
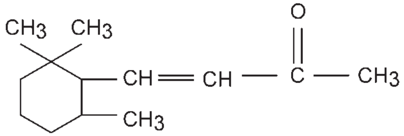
This multistage process is very complex as compared to single vaporization step and, hence, the composition of the permeate is quite different from VLE.[13] Due to high diffusivity of components in vapor phase, the desorption step is very fast and can be neglected. The overall pervaporation selectivity is mainly determined by sorption selectivity and diffusion selectivity.

Sorption

In the case of a ternary system (binary mixture and polymer), sorption is the result of the interaction between the components and also due to the interaction of the component with membrane polymer. Sorption yields the amount of solute sorbed by the membrane. Preferential sorption occurs when the membrane phase concentration is much higher than the feed liquid concentration.[14] This represents sorption selectivity.

Table 1. General properties of aroma compounds.

Compound	Structure	MW	BP (°C)	Sp. Gr.	Solubility in water (ppm)	Molar volume (cc/mol)
Linalool		154.25	198	0.86	1000	179.36
Citronellal		154.25	206	0.85	400	181.47

Menthone		154.25	207	0.9	500	171.38
β-Ionone		192.30	239	0.95	200	202.42

A number of solution theories have been developed to describe the solubility or thermodynamic interaction of a component and a polymer. Mainly two theories are used to represent sorption behavior: (i) regular solution theory; and (ii) Flory–Huggins theory. Regular solution theory has been described by Hildebrand.[15] Flory–Huggins theory has been applied in this study.

Flory–Huggins Theory[16]

According to Flory–Huggins theory the Gibbs free energy change of mixing (ΔG_{m}) of a binary mixture consisting of a solute component and polymer is given by:

$$\Delta G_{\mathrm{m}} = RT(n_{\mathrm{s}} \ln \varphi_{\mathrm{s}} + n_{\mathrm{p}} \ln \varphi_{\mathrm{p}} + \chi n_{\mathrm{s}} \varphi_{\mathrm{p}}^{2}) \tag{1}$$

where χ is Flory–Huggins interaction parameter.

From Eq. (1) it is seen that as χ increases, ΔG_{m} increases and mixing decreases.

Therefore for high affinity between polymer and solute component, the value of χ should be low.

The interaction parameter χ can be calculated from Ref.[17]

$$\chi = -\frac{\ln \varphi_{\mathrm{s}} + \varphi_{\mathrm{p}}}{\varphi_{\mathrm{p}}^{2}} \tag{2}$$

Diffusion

The sorbed component diffuses across the membrane under an activity gradient. Generally, the size of the molecule dominates the diffusion phenomenon.[17] Diffusion in a polymer is best described by Fujita's free volume theory.[18] According to this theory, the migration of the penetrant (solute component) is a sequence of unit diffusion steps during which the penetrant particle passes over a potential barrier separating one portion from the next. A unit diffusion step involves a cooperative rearrangement of the penetrant molecule and its surrounding polymer chain segments. A certain number of Vander–Waal's type interactions between the penetrant and chain segment must be broken to allow a rearrangement of local structure. The amount of energy required for this rearrangement increases with the penetrant size. If any molecule has a larger cross sectional area, it is difficult for it to pass through the polymer. Diffusion thus depends on the size of the penetrant as well as polymer segmental mobility.

Performance Parameters

Pervaporation can be characterized by performance parameters like flux and selectivity, which are indicators of the ability of the process for the extraction of a chosen component.

The permeate flux is defined as,

$$\text{flux} = \frac{W}{At} \tag{3}$$

The pervaporation selectivity is defined as,

$$\text{Selectivity} = \alpha = \frac{(w_i/w_j)_{\text{permeate}}}{(w_i/w_j)_{\text{feed}}} \tag{4}$$

There is one more parameter used to describe the pervaporation selectivity.

$$\text{Separation factor} = \beta = \frac{(w_i)_{\text{permeate}}}{(w_i)_{\text{feed}}} \tag{5}$$

For very dilute solutions where feed and permeate both contain solute concentration in ppm level, as in the case of aroma compounds in water, selectivity and separation factor are nearly equal. The overall pervaporation selectivity, α, is a product of sorption selectivity and diffusion selectivity.

Thermodynamics

A thermodynamical approach is often useful in determining the efficiency of pervaporation over distillation. Distillation process is mainly related with VLE selectivity. In order to calculate VLE selectivity for a highly diluted organic compound, 'i,' present in aqueous solution, the vapor mass fraction can be calculated by:

$$w_i^{\text{vap}} = \frac{\gamma_i x_i^{\text{liq}} M_i P_i^0(T)}{\gamma_i x_i^{\text{liq}} M_i P_i^0(T) + \gamma_j x_j^{\text{liq}} M_j P_j^0(T)} \tag{6}$$

where γ_i is activity coefficient for compound 'i' and is calculated by UNIFAC method[19] and $P^0(T)$ is saturated vapor pressure at given temperature calculated by using Lee–Kesler form of Pitzer equation, which is detailed in Reid et al.[19]

The VLE selectivity α_i^{VLE}, is given by:

$$\alpha^{\mathrm{VLE}} = \frac{(w_i^{\mathrm{vap}})/(1 - w_i^{\mathrm{vap}})}{(w_i^{\mathrm{liq}})/(1 - w_i^{\mathrm{liq}})} \tag{7}$$

The VLE separation factor is given by:

$$\beta^{\mathrm{VLE}} = \frac{w_i^{\mathrm{vap}}}{w_i^{\mathrm{liq}}} \tag{8}$$

In this way, VLE selectivity for aroma compounds can be calculated if feed conditions (feed temperature, composition of feed) are known.

EXPERIMENTAL

Materials

Elastosil LR 7600 A and B solutions were kindly supplied by Wacker Chemie, Germany, to prepare PDMS membrane. Silicalite SA-5 was supplied by Universal Oil Products (USA). The properties of silicalite filler (SA-5) are given in Table 2. *Iso*-octane solvent was procured from Merck (India) Ltd., Mumbai. Nishant Aromas, India, kindly supplied authentic samples of linalool, citronellal, menthone, and β-ionone.

Membrane Preparation

Elastosil LR 7600 A (polymer) and B (cross-linker) were mixed in 9 : 1 proportion and a 10% solution of this in *iso*-octane was prepared. It was then casted on a glass plate and cured at 80°C for 8 hr.[20] For preparation of silicalite-filled membrane (PDMS SA-5), silicalite was first homogeneously dispersed in silicone rubber. The cross-linker B was then added to this mixture and thoroughly mixed. The resulting mixture was degassed and casted on a glass plate at 80°C for 8 hr. Membranes with 20 wt% (26 vol%) loading were

Table 2. Properties of silicalite (SA-5) used supplied by Universal Oil Products, USA.

Approximate Si/Al$_2$ ratio	450
Pore diameter (Å)	6
Pore volume (cc/gm)	15

prepared. Since different membrane thicknesses were obtained by this laboratory method, fluxes reported are normalized for 100 μm membrane thickness.

Sorption Studies

Sorption studies were carried out by the membrane swelling procedure. Dry membranes of known weight were immersed in samples of pure compounds for 72 hr. These membranes were then removed and weighed after the superfluous liquid was wiped out with tissue paper. The interaction parameter of each compound for PDMS membrane calculated by using Eq. (2) and distribution coefficient between zeolite (SA-5) and PDMS calculated from sorption data of virgin PDMS and 20% silicalite membrane are given in Table 3. The distribution coefficient between PDMS and zeolite is defined as,

$$K_Z = \frac{C_Z}{C_M} \tag{9}$$

The term K_Z represents adsorbability of the zeolite. Higher value of K_Z means good adsorbability. Generally the value of K_Z for an organic compound and silicalite depends upon hydrophobicity of the organic compounds. Thus, the more hydrophobic the compound, the higher is the value of K_Z.

Permeation Studies

Pervaporation experiments were carried out in a batch-stirred cell operated under vacuum as shown in Fig. 1.[21] The cell had two flanged compartments. The upper compartment, containing liquid feed with a capacity of 550 mL was provided with an outer jacket for temperature control. The lower compartment was connected to a vacuum pump through a liquid nitrogen trap. The membrane was sandwiched between the two compartments on a porous stainless steel

Table 3. Interaction parameter and distribution coefficient values of the aroma compounds.

Compound	χ_{PDMS}	$K_Z = C_Z/C_M$
Linalool	0.933	1.2176
Citronellal	1.042	1.0827
Menthone	0.636	1.7843
β-Ionone	1.4	1.0405

Note: χ_{PDMS}, interaction parameter of PDMS membrane.

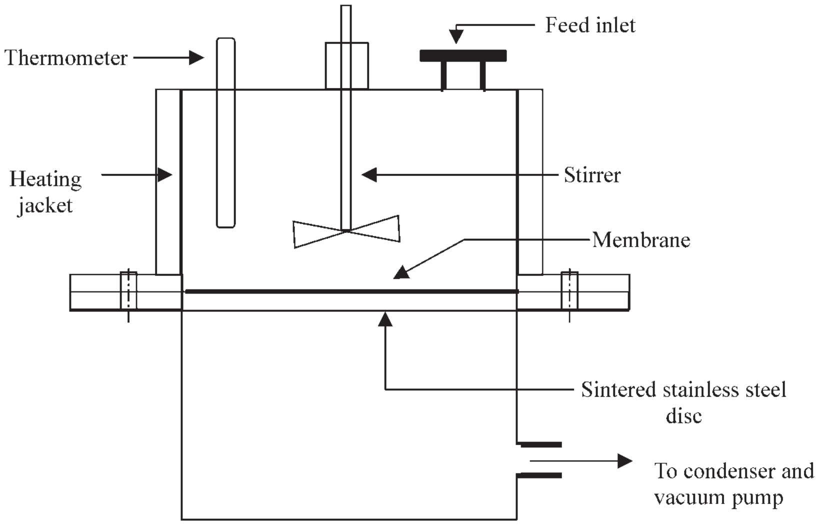


Figure 1. Experimental set-up for pervaporative separation.

sintered disc and sealed with O-ring. The permeate side pressure was maintained constant at 2 mmHg. Effective membrane separation area was 38.5 cm^2. The vapor permeate was condensed by using a liquid nitrogen trap.

Analysis

The feed and permeate concentrations of all the binary mixtures were analyzed by measuring the absorbance of UV–VIS light in these solutions by Chemito 2100 UV–VIS spectrophotometer at corresponding wavelengths (Table 4).

Table 4. The wavelength values corresponding to maximum absorbance for the aroma compounds.

Compound	Wavelength (nm)
Linalool	205
Citronellal	480[a]
Menthone	480[a]
β-Ionone	308

[a] Wavelength found from colorimetric method.[22]

Calibration curves (concentration vs. absorbance) for linalool and β-ionone were made. Since the carbonyl compounds (citronellal and menthone) do not give maximum absorbance in UV range, their analysis was done by colorimetric method.[22] In this method a color complex was formed whose absorbance was measured in visible range. Then calibration curves for these compounds were made in the visible range.

A two-phase mixture was obtained as a permeate. This permeate was further diluted to obtain a homogeneous mixture. The concentration of a solute in the permeate was found out from the calibration curve.

Permeation Through Zeolite-filled Membrane

When suitable zeolites are incorporated in the membrane they can act as reservoirs for the preferentially adsorbed species. Netke et al.[23] presented a model for diffusion of solutes through zeolite-filled membrane. In this model, total flux across zeolite-filled membrane is given as:

$$J = -\frac{dC_M}{dx}(D_{MM} + D_{MZ}(1-\theta)) - \frac{dC_Z}{dx}\left(\frac{C_M}{C^*}D_{MZ} + D_{ZM} + D_Z + D_{ZZ}\right) \tag{10}$$

The equation for integral permeability is given by:

$$P = k_M\left(D_{MM} + ([D_{ZM} + D_{ZZ} + D_Z]k_Z C^* - D_{MZ})(1-\theta_0) - 2D_{MZ}\left(\frac{(1-\theta_0)}{\theta_0}\right)\ln(1-\theta_0)\right) \tag{11}$$

The permeability of filled membrane given by Eq. (11) has the following asymptotes:

when $\theta_0 \ll 1$ (poor adsorption)

$$P = k_M[D_{MM} + (D_{ZM} + D_{ZZ} + D_Z)k_Z C^* + D_{MZ}] \tag{12}$$

when $\theta_0 \sim 1$ (strong adsorption)

$$P = k_M(D_{MM}) \tag{13}$$

Equation (12) shows an increase in P over that for the membrane alone if the terms other than D_{MM} are of the same order of D_{MM}. The permeability of the filled membrane is governed by k_M, k_Z, and various diffusivity values. Characteristic constants for the solute-membrane-filler system under consideration are k_M and k_Z because $D_Z \ll D_{MM}$, hence the diffusion in the pores of the zeolite is much slower than that in the membrane polymer. Further, the presence of zeolite particles creates a tortuous and longer diffusion path. The overall effect is, therefore, a lower organic flux in the presence of the zeolite. The diffusivities are, however, not only characteristic system properties but are also likely to vary with the filler content and nature of adsorption (strong or weak).

Netke et al.[23] have shown that the selectivity for the organic–aqueous system permeating in zeolite-filled membrane increases with the increase in adsorbability of the organic with respect to the zeolite. A detailed explanation for the same is also given by Netke et al.[23]

RESULTS AND DISCUSSION

In pervaporation of aroma compounds from their aqueous solutions, concentration polarization in the liquid film can develop. Under these conditions the true membrane permeation is masked by the external diffusion (concentration polarization) resistance. In the present case, the upstream liquid feed compartment of the cell was provided with an axial downflow turbine. The flow generated was directed towards the membrane surface. This flow swept the membrane surface thereby assisting in elimination of concentration polarization. Preliminary experiments were carried out at varying speed of agitation and flux values were determined. It was found that starting with low speed (6.67 rev/sec) the flux values increased and then became constant at and above 20 rev/sec for the 100-μm thick membrane used as shown in Fig. 2. In the experimental data reported in this work all the experiments were carried out at 21.67 rev/sec.

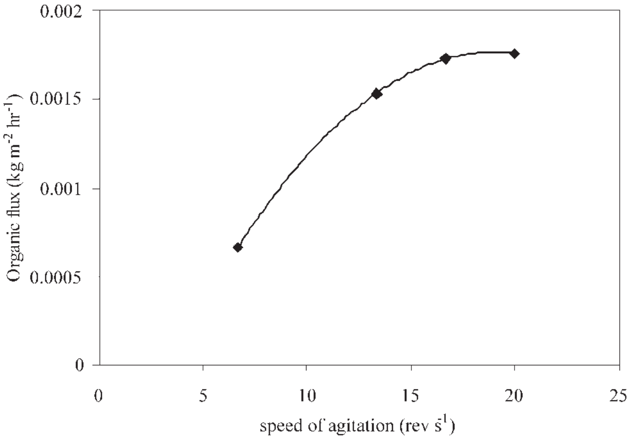


Figure 2. Variation of organic flux with speed of agitation.

As a general rule, it can be assumed that the more hydrophobic (or apolar) the organic compound is, the better is the efficiency of an organophilic pervaporation membrane. Nevertheless, the behavior of the pervaporation membranes cannot be generalized regardless of the nature (composition and structure) of the aroma compounds to be extracted.

In the present study, pervaporation performance of aqueous solutions of aroma compounds having different functional groups were studied. The binary solution of each compound studied has concentrations less than its solubility limit in water.

The pervaporation study of different aroma compounds is divided as per their functional group in the following manner.

Alcohol

Linalool is a terpene alcohol with open structure (Table 1). Its low value of interaction parameter value in virgin PDMS as well as high distribution coefficient between PDMS and zeolite (Table 3) indicates sorption of linalool in both virgin PDMS and in zeolite is high. Since linalool has an open structure, its diffusion coefficient is likely to be high. The organic fluxes were higher compared to other aroma compounds. Figure 3 shows the

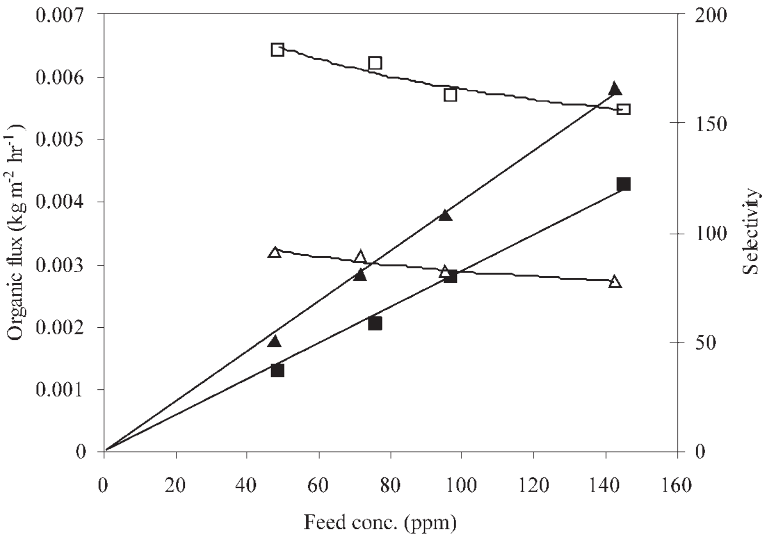


Figure 3. Variation of flux and selectivity of linalool with its feed concentration in PDMS and PDMS SA-5 membrane. *Key:* △, PDMS; □: PDMS SA-5; filled symbols: organic flux; hollow symbols: selectivity.

comparison of organic fluxes for both the membranes. It can be been seen that the organic flux increases with feed concentration. But there was a decrease in organic flux in PDMS SA-5 membrane as compared to PDMS membrane for the same feed concentration. This is because of strong binding (higher K_Z) of organic molecule to the silicalite adsorbent and the longer diffusional path in zeolite-filled membrane, which decreases its permeation through the membrane.[23] Figure 3 also shows the comparison of selectivity for the two membranes. It has been found that selectivity decreases with feed concentration of the organic compound. On the other hand, the selectivity for PDMS SA-5 membrane was almost double that of PDMS membrane because of more sorption in case of PDMS SA-5 membrane as compared to PDMS membrane as explained by Netke et al.[23] In Fig. 4 the permeate concentration with feed concentration of linalool for both the membranes and VLE are compared. It can be observed that PDMS membrane yields marginally higher permeate concentration while PDMS SA-5 gives much higher permeate concentration than that of VLE. The comparison of pervaporation selectivity with VLE selectivity of different aroma compounds at the same feed concentration is shown in Fig. 5, which indicates that even though VLE selectivity of linalool is high [due to high γ and average $P^0(T)$], overall pervaporation selectivity is greater because of high sorption and good diffusion.

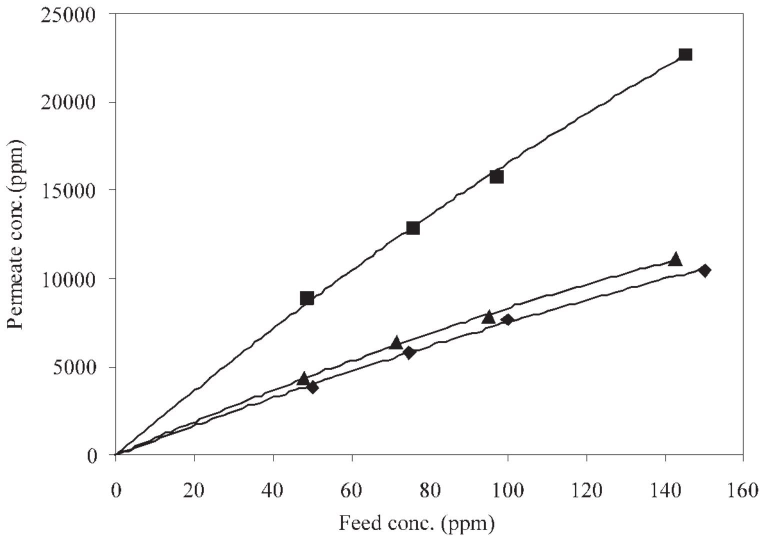


Figure 4. Variation of permeate concentration of linalool with its feed concentration. *Key:* ▲, PDMS; ■, PDMS SA-5; ◆, VLE.

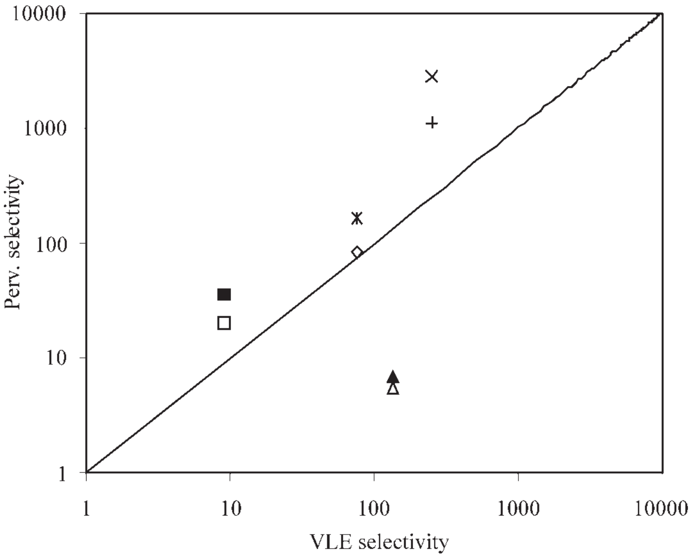


Figure 5. Comparison between pervaporation selectivity and VLE selectivity. *Key:* □, Citronellal (PDMS); ■, citronellal (PDMS SA-5); ◇, linalool (PDMS); *, linalool (PDMS SA-5); △, β-ionone (PDMS); ▲, β-ionone (PDMS SA-5); +, menthone (PDMS); ×, menthone (PDMS SA-5).

Aldehyde

Citronellal is a terpene aldehyde also having an open structure (Table 1). Its higher interaction parameter value for virgin PDMS (Table 3) indicates less sorption than that of linalool. The distribution coefficient between PDMS and SA-5 is less as compared to linalool (Table 3). When organic fluxes and selectivity of both the membranes were compared as shown in Fig. 6, it was found that the difference between organic flux and selectivity of PDMS and PDMS SA-5 membranes was less as compared to linalool because of its lower sorption in the zeolite (lower K_Z). Figure 7 shows comparison of permeate concentration of citronellal with feed concentration for both the membranes and VLE. It can be observed that because of low VLE permeate concentration both membranes show permeate concentration well above that for VLE. Because of low VLE selectivity [low γ and $P^0(T)$] for citronellal, both the membranes give greater pervaporation selectivity than VLE selectivity as shown in Fig. 5.

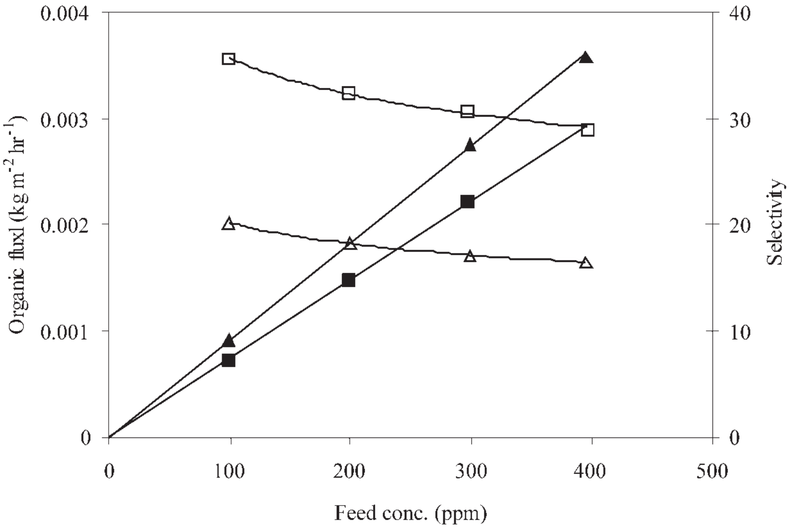


Figure 6. Variation of flux and selectivity of citronellal with its feed concentration in PDMS and PDMS SA-5 membrane. *Key:* △, PDMS; □, PDMS SA-5; filled symbols: organic flux; hollow symbols: selectivity.

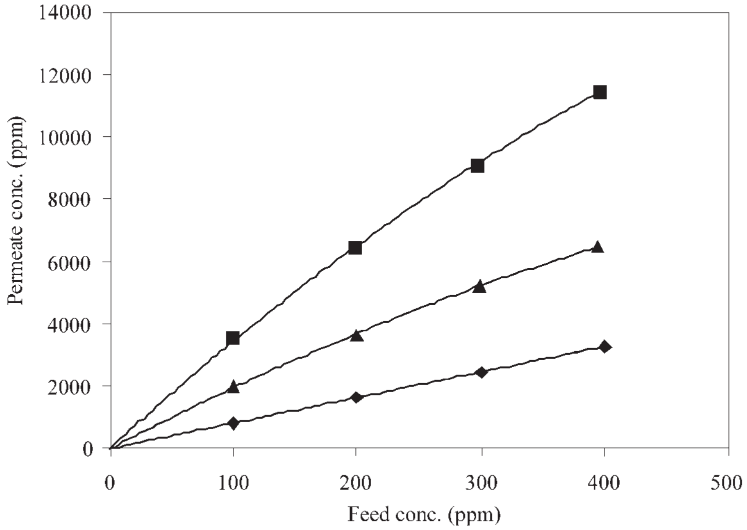


Figure 7. Variation of permeate concentration of citronellal with its feed concentration. *Key:* ▲, PDMS; ■, PDMS SA-5; ◆, VLE.

Ketone

In this work the two types of ketones studied are menthone and β-ionone. Menthone is a terpene ketone having ring structure (Table 1). Due to its very high sorption (low χ value), the pervaporation performance of menthone was found to be best among the compounds studied. The distribution coefficient between PDMS and SA-5 for menthone is highest among the compounds studied (Table 3). In Fig. 8 organic fluxes of both the membranes are compared. It was observed that there was a drastic decrease in organic fluxes in the case of PDMS SA-5 membrane due to strong binding of menthone molecule in adsorbent filler (highest K_Z) and longer diffusion path as described by Netke et al.[23] Also there was about a three times increase in selectivity in PDMS SA-5 membrane when the two membranes were compared, as shown in Fig. 8. The organic fluxes were less than that of linalool because of poor diffusion of menthone molecule having a larger cross sectional area (because of ring structure). In Fig. 9 comparison of permeate concentration with feed concentration of menthone for both the membranes and VLE is given. Due to high sorption, both the membranes give permeate concentration greater than that of VLE. Although menthone has high VLE selectivity [high γ

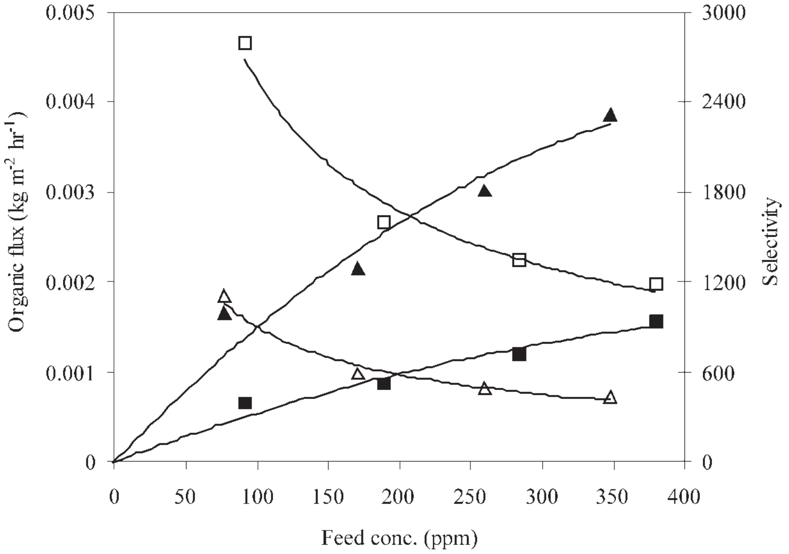


Figure 8. Variation of flux and selectivity of menthone with its feed concentration in PDMS and PDMS SA-5 membrane. *Key:* △, PDMS; □, PDMS SA-5; filled symbols: organic flux; hollow symbols: selectivity.

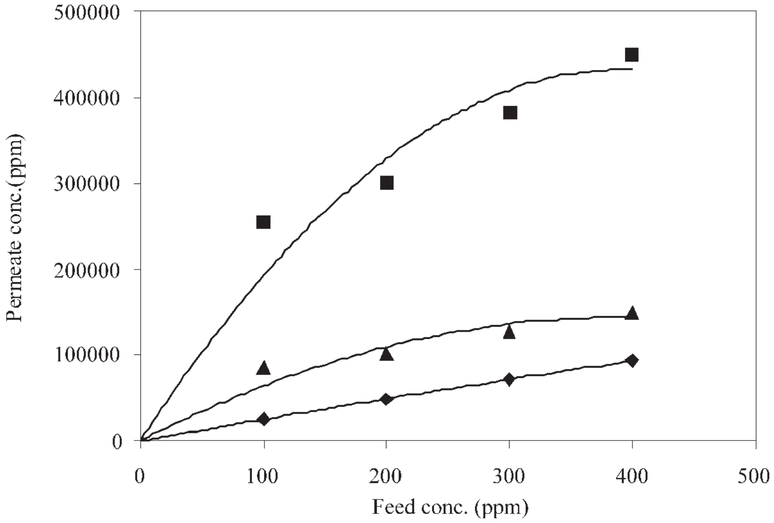


Figure 9. Variation of permeate concentration of menthone with its feed concentration. *Key:* ▲, PDMS; ■, PDMS SA-5; ◆, VLE.

and $P^0(T)$], it has much less than overall pervaporation selectivity because of very high sorption as shown in Fig. 5.

β-Ionone is a sequiterpene also having a ring structure (greater cross sectional area). Its high value of χ in virgin PDMS and low distribution coefficient between PDMS and zeolite (Table 3) indicates low sorption in PDMS as well as in the zeolite. Also, due to larger cross sectional area, its diffusion through the membrane is very slow. Hence, there were marginal changes in organic fluxes as well as pervaporation selectivity when the two membranes were compared, as shown in Fig. 10. Because of very high γ, its VLE permeate concentration and VLE selectivity are high. Figure 11 shows differences in permeate concentrations for both the membranes and VLE. It is observed that VLE permeate concentrations of β-ionone are much greater than that for both the membranes. From Fig. 5 it is seen that the overall pervaporation selectivity of both the membranes is less than that of VLE selectivity. The organic fluxes and pervaporation selectivity for β-ionone are relatively much lower as compared to other aroma compounds because of very low sorption as well as poor diffusion.

In order to show relationship between transport properties and thermodynamics, the plots of organic flux vs. fugacity for different aroma compounds

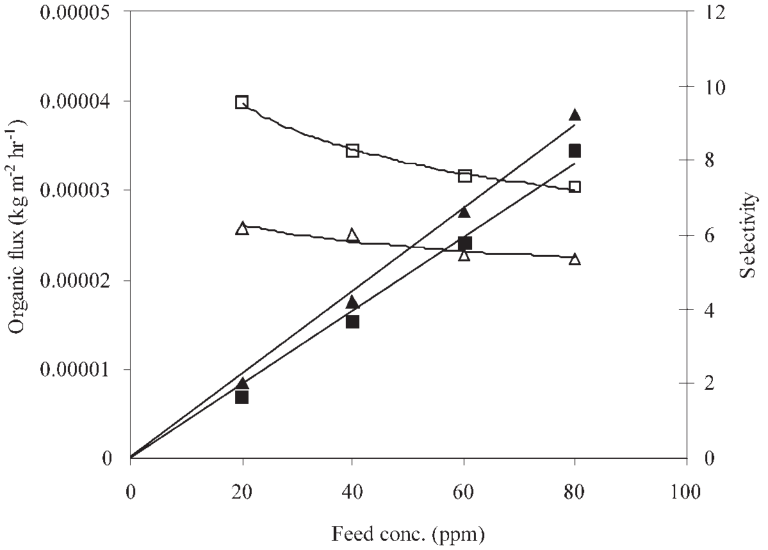


Figure 10. Variation of flux and selectivity of β-ionone with its feed concentration in PDMS and PDMS SA-5 membrane. *Key:* △, PDMS; □, PDMS SA-5; filled symbols: organic flux; hollow symbols: selectivity.

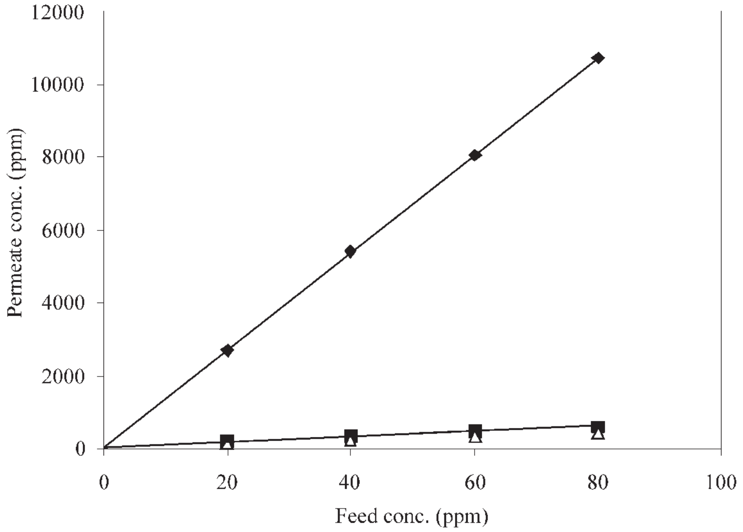


Figure 11. Variation of permeate concentration of β-ionone with its feed concentration. *Key:* △, PDMS; ■, PDMS SA-5; ◆, VLE.

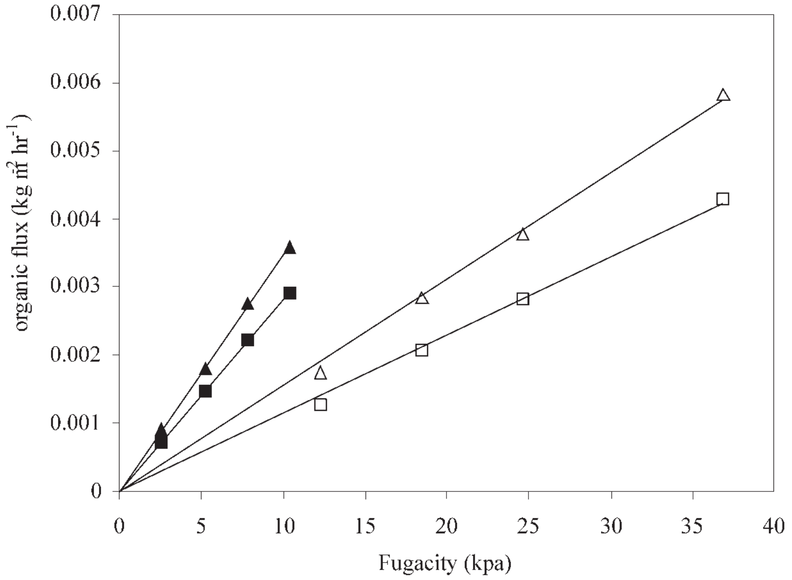


Figure 12. Variation of flux of linalool and citronellal with fugacity. *Key:* △, PDMS; □, PDMS SA-5; filled symbols: citronellal; hollow symbols: linalool.

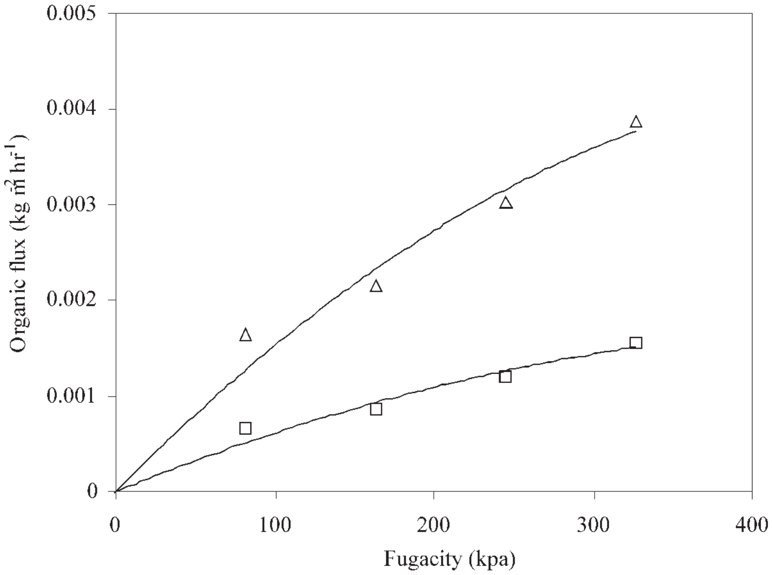


Figure 13. Variation of flux of menthone with fugacity. *Key:* △, PDMS; □, PDMS SA-5.

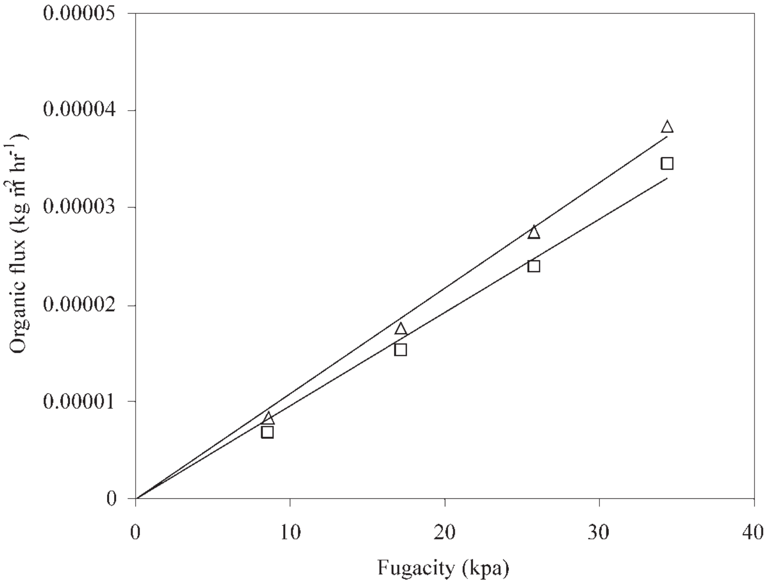


Figure 14. Variation of flux of β-ionone with fugacity. *Key:* △, PDMS; □, PDMS SA-5.

are shown in the Figs. 12–14. In these plots chemical potential driving force fugacity is calculated by using γ at infinite dilution as an independent variable.

CONCLUSION

Pervaporation performances of aroma compounds are very much dependent on type of membrane as well as the nature of the aroma compounds. Among the components studied menthone yielded highest overall pervaporation selectivity because of high sorption while β-ionone, having low sorption, yielded lowest overall pervaporation selectivity. Further, when the compound has a strong binding of organic molecule with adsorbent filler there is a decrease in organic flux in filled membrane (PDMS SA-5) as compared to unfilled (PDMS) membrane. The pervaporation selectivity for filled membrane is also greater than unfilled membrane. The nature of aroma compound also affects pervaporation performances as in the case of β-ionone. Here because of the larger cross sectional area, diffusion of β-ionone through the membrane is poor resulting in less overall pervaporation selectivity as compared to other aroma compounds.

Most of the aroma compounds give good pervaporation selectivity using PDMS and PDMS SA-5 membranes. It has been observed that the aroma compounds having high sorption always yield a higher overall pervaporation selectivity than VLE selectivity.

NOTATIONS

ΔG_m	Gibbs free energy change of mixing
A	effective membrane area (m^2)
C	concentration (kg/m^3)
C^*	saturation concentration in Langmuir isotherm (kg/m^3)
D	diffusivity (m^2/s)
J	organic flux through pervaporation membrane ($kg/m^2\,hr$)
k_M	constant for solute sorption (kg solute/kg dry membrane)
k_Z	constant in Langmuir isotherm
K_Z	distribution coefficient between PDMS and SA-5
M	molecular weight
n	number of moles
P	permeability (m^2/s)(kg solute/kg dry membrane)
$P^0(T)$	saturated vapor pressure at a given temperature, T
R	universal gas constant
t	time
T	absolute temperature (K)
w	weight fraction
W	weight of permeate (kg)
x	liquid mole fraction
φ	volume fraction
θ	fraction of zeolite sites occupied at a given location in the membrane
θ_0	fraction of zeolite sites occupied at feed side location
α	selectivity
β	separation factor
γ	activity coefficient
χ	interaction parameter

Subscripts

i	desired component
p	polymer
s	solute component

M	membrane phase
Z	zeolite phase
feed	refers to feed
liq	liquid phase
permeate	refers to permeate
vap	vapor phase
VLE	vapor–liquid equilibrium

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