Improved conversion to ethyl acetate through removal of water of esterification by membrane pervaporation

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Crosslinked poly (vinyl alcohol) (PVA) membranes were prepared using maleic acid (MA) and citric acid (CA) as crosslinking agents. First the water separation ability of the crosslinked membranes from aqueous acetic acid (10-90% acetic acid) and ethanol (20-80% ethanol) solutions at 30 to 90°C was tested by pervaporation. The effects of temperature, feed concentration, crosslinker type on pervaporation characteristics were investigated. The PVAMA membrane showed separation factors of 1.9-16.2 and fluxes of 4.26-38.48 kg/m² h and PVACA membrane showed separation factors of 3.8-33 and fluxes of 2.17-480.8 kg/m² h for acetic acid-water separation. In case of ethanol-water separation, the PVAMA membrane showed separation factors of 0.7-17.9 and fluxes of 2.5-36.2 kg/m² h and PVACA membrane showed separation factors of 1.4-41.9 and fluxes of 3.8-359.7 kg/m² h. PVACA membrane has shown high separation efficiency of water than that of PVAMA membrane. Having high selectivity and good permeating ability for water both the membranes were then used to remove water of esterification from reaction of acetic acid with ethanol by a batch pervaporation. Compared to around 60% conversion in conventional esterification, up to 98% conversion to ethyl acetate was achieved from ethanol and acetic acid when water by product was removed by pervaporation using the prepared membranes at same temperature.

Keywords: Pervaporation, PVA membrane, Esterification, Flux

IPC Code (s): B01D17/00, B01D61/36

Pervaporation is a membrane separation process as well as purification technique used for the separation of desired component from a liquid/liquid mixture. This technique consists of permeation of components into a membrane followed by evaporation on the permeate side and operates on the principle of selective permeation of a liquid component through a membrane.

Dehydration of acetic acid by membrane pervaporation has not received much attention as the membranes often fail during operation due to poor strength and stability. Membranes with high degree of crosslinking are needed for the dehydration of acetic acid. Purification of organic solvents, which forms azeotrope, such as methanol, ethanol, and isopropanol containing a small amount of water is of vital significance in the area of organic synthesis. These solvents are important as they are used in a large scale in chemical industries as well as in pharmaceutical laboratories. Their separation requires the use of energy intensive processes such as extractive and azeotropic distillation, vacuum distillation and solvent extraction etc. Pervaporation offers more economical alternative for such difficult separation of components from their mixtures. Since the performance of a pervaporation process depends on the intrinsic properties of the polymer membrane, the preparation of novel membranes with good pervaporation performance is very important. Although PVA has some unique characteristics such as good chemical stability, film forming ability and high hydrophilicity but one of its limitations is its high solubility in water. So in order to get a stable membrane it must be insolubilized by crosslinking. Huang and Yeom separated ethanol-water and acetic acid-water mixtures using amic acid crosslinked PVA membrane. The PVA membrane crosslinked with 12-wt% amic acid gave separation factors of 13-42 and permeation rates of 0.079-2.285 kg/m².h depending on the operating temperatures and feed composition. Yeom and Lee prepared PVA membranes crosslinked with glutaraldeyde and got a separation factor of 120-420 and flux of 0.029-0.263 kg/m².h. Toti and Aminabhavi used sodium alginate and modified sodium alginate membranes to separate water-acetic acid and water-isopropanol mixtures. Of all the membranes studied, sodium alginate membranes containing 10-wt% of polyethylene glycol and 5-wt% of PVA gave the highest selectivity to water, i.e., ~40 for water-acetic acid mixture.
Aminabhavi and Naik\textsuperscript{14} prepared PVA-g-poly(acrylamide) copolymeric membrane to separate acetic acid-water mixtures. They got a flux of 0.008-0.042 kg/m\textsuperscript{2}.h and separation factor of 5.36 to 8.53 at a feed concentration of 10% water depending on the polyacrylamide loading in the membrane. Huang and Rhim\textsuperscript{15} used maleic acid for crosslinking of PVA and separated water-acetic acid mixtures. They got a flux of 0.180 kg/m\textsuperscript{2}.h and a separation factor of 7.8 at 25\textdegree C from a feed containing 10% water.

Esterification of carboxylic acids and alcohols are well known examples of equilibrium-limited reactions, which is a challenging problem in product purification. Membrane pervaporation is a promising approach\textsuperscript{16} to overcome such problem. Since the separation involves selective permeation of a component from a mixture, membranes can help to enhance the conversion of thermodynamically or kinetically limited reactions through controlled removal of one or more product species from the reaction mixture\textsuperscript{17}. Jennings and Binnings\textsuperscript{18} proposed this concept of removing by product from reaction mixtures. Esterification of oleic acid and ethanol\textsuperscript{19}, propionic acid and propanol\textsuperscript{20}, oleic acid and butanol\textsuperscript{21}, valeric acid and ethanol\textsuperscript{22} were carried out in presence of various acids or lipases catalysts. Most of the researchers carried out esterification applying pervaporation to remove water using inorganic membranes\textsuperscript{23-25}. In most of the ethanol and acetic acid reactions people used zeolite membranes or zeolite filled polymeric membranes. Zeolite based membranes are unstable to acids and are not applied to systems in direct contact with acids\textsuperscript{26}. Only a few reports are available on the use of polymer membranes to remove water of esterification by pervaporation\textsuperscript{27,28}. Also none has used maleic acid (MA) and citric acid (CA) crosslinked PVA membranes in those esterifications to remove water by pervaporation.

The main objective of this investigation, therefore, is the development of two stable crosslinked PVA membranes: one is MA crosslinked PVA (PVAMA) and the other is CA crosslinked PVA (PVACA). To judge the suitability for water permeation, the prepared membranes were first used to dehydrate ethanol and acetic acid separately. The membranes were then used for the removal of water of esterification of ethanol and acetic acid by pervaporation.

### Experimental Procedure

#### Materials

Polyvinyl alcohol (Mol. Wt. 1,00,000) was purchased from Fluka AG. Maleic acid (MA) was procured from Loba Chemie, India. Citric acid (CA), acetic acid, sulfuric acid and ethanol were purchased from E. Merck, India.

#### Preparation of crosslinked PVA membranes

PVA powder was dissolved in distilled water by heating at 95\textdegree C for 5 h to produce a clear 10 wt.% solution. For crosslinking of PVA, calculated amounts of crosslinking agents, viz., CA and MA were dissolved in PVA solution along with one drop of concentrated sulphuric acid catalyst. After the membrane was cast on a flat base petridish, water was allowed to evaporate at 30\textdegree C. Then the membrane was subjected to curing in an oven at 70\textdegree C for 1 h. The amount of crosslinking agent was optimized to achieve maximum degree of crosslinking of PVA. In this investigation 19-mole% crosslinker loading to PVA could be done in both the cases.

#### Infrared spectroscopic analysis

The surface chemical structures of the crosslinked PVA membranes were analyzed in ATR mode by a FTIR spectrophotometer (Thermo Nicolet, Nexus 870, using a zinc selenide crystal and OMNIC software).

#### X-ray diffraction (XRD) analysis

To know the extent of crystallinity of prepared crosslinked PVA membranes XRD study was done using Philips PW 1729 XRD analyzer for the range 2\theta = 10\textdegree-40\textdegree using Cu-K\textalpha as target.

#### Contact angle measurement

In order to measure the surface wettability of the membrane, contact angle of PVA, PVAMA and PVACA membrane surfaces were measured by Rame Goniometer (model 100-00-230).

#### Tensile property measurement

Tensile strength and elongation at break of crosslinked PVA membranes were measured using Hounsfield H10KS Universal Testing Machine (UTM). According to ASTM specifications, samples of rectangular strips of 6 mm width were cut out from the polymer membrane. The strips were well gripped using thick paper for the measurement of tensile strength at a grip separation speed of 50 mm/min.
Pervaporation

The pervaporation cell was assembled from two cylindrical half-cells made of stainless steel fastened together by nuts and bolts. The membrane was supported on a perforated stainless steel plate placed at the junction of two cells. The feed temperature was maintained by circulating hot water through the cell jacket. For all measurements, the downstream pressure was maintained at 666.61 Pa (5 mm Hg) by applying vacuum. The permeate vapor was collected in a glass condenser suspended inside a cryogenic trap kept at -40°C. The experiment was run for 3 h with no change in flux with time, confirming the steady-state operation. The effective membrane area for pervaporation was 0.002826 m². First the water removal capacity of the membranes from aqueous acetic acid and ethanol by pervaporation was evaluated. Then the progress of esterification reaction between acetic acid and ethanol at different temperatures, viz., 70, 80 and 90°C was monitored by removal of water of esterification by pervaporation using the prepared membranes. The condensed permeate was warmed up to ambient temperature and then collected for analysis by acid base titration. The change in feed composition after pervaporation run was also analyzed by quantitative acid base titration method. All the pervaporation runs were repeated thrice for each of the feed compositions studied with 0.2-0.5% variations in flux data and the average results are reported.

The flux \( J \) can be calculated from the weight of permeate collected after pervaporation run using Eq. (1).

\[
J = \frac{Q}{A \cdot t}
\]  

where, \( Q \) is total weight of permeates collected in time \( t \) and \( A \) is effective membrane area.

The separation factor \( \alpha_{ij} \) is defined by,

\[
\alpha_{ij} = \frac{y_j/x_j}{x_i/y_i}
\]  

where, \( x_i \) and \( x_j \) are the weight fractions of the component in feed and \( y_j \) and \( y_i \) are the weight fractions of those in the permeate respectively. Component i was water in this experiment, separation effectiveness was characterized by separation selectivity of water compared to the other components.

The progress of esterification reaction was monitored by estimating acetic acid using Eq. (3).

\[
\%\text{Ethyl acetate formed} = \frac{G - (G_i + G_j)}{G} \times 100\%
\]

where, \( G \) is the initial mass of acetic acid (kg), \( G_i \) is the mass of the residual acid in the feed mixture (kg), and \( G_j \) is the mass of the permeated acid (kg).

Results and Discussion

Membrane synthesis

As the separation performance of aqueous ethanol and acetic acid depends upon the morphology and chemical composition of membrane, two types of crosslinked PVA membrane of hydrophilic nature were synthesized in this investigation. Such a membrane with adequate crosslink density is expected to provide selective permeation of water from its mixture with organics in feed. In this investigation, crosslinking of PVA with either MA or CA was done by esterification of some of hydroxyl group of PVA with the carboxyl groups of MA or CA. Since such crosslinking by esterification is a very slow reaction, which was done at solid phase at 70°C, around 28-30 mole % crosslinker was used to obtain a substantial amount of crosslinking. After curing the unreacted crosslinker was leached out from the membrane by boiling in water till the leached water became acid free. From a difference in weight of the membrane before and after curing and washing it was found that the membrane retained 19 mole% crosslinker attached to polymer chain (Table 1). Out of this 19-mole% crosslinking agent, one part was consumed in crosslink bond formation and the other part is linked to polymer chain through an ester bond leaving one

<table>
<thead>
<tr>
<th>Membrane</th>
<th>Crosslinker (mole %)</th>
<th>% Crystallinity</th>
<th>Thickness (μm)</th>
<th>Contact angle (degree)</th>
<th>Tensile strength (MPa)</th>
<th>Elongation at break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVA</td>
<td>0</td>
<td>10.35</td>
<td>40</td>
<td>49.5</td>
<td>41.7</td>
<td>11.7</td>
</tr>
<tr>
<td>PVACA</td>
<td>19</td>
<td>9.52</td>
<td>40</td>
<td>58.5</td>
<td>43.4</td>
<td>13.1</td>
</tr>
<tr>
<td>PVAMA</td>
<td>0</td>
<td>7.46</td>
<td>40</td>
<td>62.5</td>
<td>44.2</td>
<td>12.6</td>
</tr>
</tbody>
</table>
carboxyl group free. However, the amount of crosslinker fully participated in crosslink bond formation was not assessed in this investigation. That the cured membrane contained free carboxyl group of partially linked crosslinking agent was ascertained by FTIR spectroscopy discussed later.

FTIR (ATR) spectral analysis
For structural characterization, ATR spectra of both uncrosslinked and crosslinked PVA membranes were taken (Fig. 1). The occurrence of crosslinking is evident from the IR spectra. The broad peak at 3300-3400 cm\(^{-1}\) corresponds to hydroxyl groups in PVA. A decrease in the intensity of this peak for crosslinked PVA membranes was due to reaction of hydroxyl group with the carboxyl group of maleic acid or citric acid. A strong peak at 1700–1750 cm\(^{-1}\) in the ATR spectra of crosslinked PVA membranes appeared due to free and hydrogen bonded carbonyl groups of ester crosslink bond. In crosslinked PVAMA there is a strong C=O stretching vibration at 1710 cm\(^{-1}\) which indicated the presence of an ester group. Similar bands also appeared for crosslinked PVACA membrane. The strong C=O stretching vibration at 1715 cm\(^{-1}\) in PVACA proves the crosslinking of PVA through esterification reaction between –OH groups of PVA and –COOH groups of CA. It was mentioned in the previous section that both the carboxyl groups of all the CA or MA molecules would not react to form ester crosslink bonds rather one carboxyl group of few CA or MA molecules would remain as free pendant group. This aspect was corroborated by FTIR spectroscopy, which showed the appearance of a peak at 2570 cm\(^{-1}\) due to O-H stretching of free COOH group.

XRD analysis
Due to crystalline nature, PVA molecules in the membrane form compact morphology and dense system, which cannot be suitable for higher pervaporation flux. The PVA membrane was, therefore, crosslinked to serve two major purposes: one is to make the membrane insoluble in aqueous medium and the other is to decrease the crystalline order of PVA molecules so that higher permeate flux can be obtained. To study the effect of crosslinking on the morphology of the PVA membrane, XRD analysis was done. XRD patterns of both uncrosslinked and crosslinked PVA membranes are shown in Fig. 2. Percent crystallinity of the PVA, PVAMA and PVACA membranes was calculated and shown in Table 1. It is observed that after crosslinking the crystallinity of PVA is decreased.

Contact angle measurement
Contact angle gives a measure of hydrophilicity or hydrophobicity of a material. With decrease in the contact angle hydrophilicity of a membrane is increased. Due to water-soluble nature PVA as such is highly hydrophilic. From contact angle measurement it is observed that the hydrophilicity of PVA membrane is decreased after crosslinking with either MA or CA. The PVACA membrane is more hydrophilic than PVAMA membrane (Table 1). However, the observed contact angle data of the crosslinked membranes indicate their adequate surface wetting nature required for selective permeation of water through them.

Tensile property
In order to judge the ability of the prepared membrane to withstand handling and mechanical
stress during the pervaporation operation the tensile property of the prepared membranes was measured. The mechanical properties of the synthesized crosslinked PVA membranes such as tensile strength and elongation at break were measured and the results are shown in Table 1. It is seen from the data that the membranes are quite strong mechanically.

Pervaporation separation of water- acetic acid and water ethanol mixture

Before subjecting the crosslinked PVA membranes for removal of water of esterification, first their pervaporation parameters were evaluated for separation of water from aqueous acetic acid and ethanol solutions. Tables 2 and 3 show the effect of feed composition, temperature and crosslinker type on pervaporation separation characteristics of water-acetic acid and water-ethanol mixtures. The effect of feed composition in the range of 10-90 wt.% acetic acid in water and 20-80% ethanol in water on the permeation flux and separation factor was investigated. The results are shown for three temperatures 30, 45 and 90°C and for two types of membranes PVAMA and PVACA. It is clear that when the water concentration in feed increases the permeation rate or flux increases and separation factor decreases. Similar results were obtained for both PVAMA and PVACA membranes.

The permeation flux of water is higher than that of acetic acid/ethanol regardless of feed composition (Tables 2-3). As the water in the feed is increased the membrane is more swollen; so the flexibility of polymer chains increases and the energy required for diffusive transport through membrane decreases. Another important factor is molecular size. Acetic acid has larger molecular size than water and ethanol molecules. As the amount of ethanol/acetic acid increases in the feed mixture it becomes difficult for ethanol/acetic acid molecule to diffuse through the swollen membrane. Again at higher concentration acetic acid molecules are dimerised, but ethanol molecule cannot do so. There is strong intramolecular hydrogen bonding in acetic acid, but there is no scope of forming intramolecular hydrogen bonding in case

<table>
<thead>
<tr>
<th>Temp (°C)</th>
<th>PVAMA</th>
<th></th>
<th>PVACA</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed</td>
<td>Permeate</td>
<td>% Water in Acetic acid</td>
<td>Water</td>
<td>% Water in Acetic acid</td>
</tr>
<tr>
<td>30</td>
<td></td>
<td>98.3</td>
<td>6.6</td>
<td>33.3</td>
</tr>
<tr>
<td>45</td>
<td></td>
<td>98.5</td>
<td>6.7</td>
<td>34.4</td>
</tr>
<tr>
<td>90</td>
<td></td>
<td>99.1</td>
<td>12.2</td>
<td>38.5</td>
</tr>
</tbody>
</table>

Table 2 — Pervaporation separation of water from aqueous solution of acetic acid using 2.83×10⁻³ m² PVAMA and PVACA membranes of thickness 40 μm after 3 h continuous run.
of ethanol. So flux and separation factor of water are higher at high acetic acid concentration than that of ethanol.

The above pervaporation separation performance was studied for three temperatures 30, 45, and 90°C (Tables 2-3). Since the esterification reaction between ethanol and acetic acid occurs at 70°C, it is required to check the membrane behavior with increase in temperature. As the temperature increases the permeation flux increases. With increase in temperature the free volume of the polymer membrane increases which facilitates the transport of permeants through the membrane. It is clear from the data that at higher temperature membranes have better separation efficiency of water.

From XRD pattern of crosslinked PVA membrane it is clear that peak intensity of PVAMA membrane is less compared to uncrosslinked PVA and PVACA. This is a strong indication that the crystallinity of the PVAMA is less than that of PVACA. Again from contact angle data it is clear that PVACA membrane is more hydrophilic than PVAMA membrane, i.e., PVACA membrane has better water absorbing ability than PVAMA membrane. For this reason PVACA membrane shows more permeation of water than that of PVAMA membrane (Tables 2-3). Another reason for more permeation of water through PVACA membrane is the bigger size of citric acid molecule as crosslinking agent, which contributes higher free volume to the membrane. Therefore, small water molecules can pass easily through PVACA membrane compared to acetic acid and ethanol due to their steric hindrances. These results indicate the suitability of the crosslinked PVA membranes for selective removal of water of esterification of acetic acid and ethanol.

Table 3 — Pervaporation separation of water from aqueous solution of ethanol using $2.83 \times 10^{-3}$ m² PVAMA and PVACA membranes of thickness 40 μm after 3 h continuous run.

<table>
<thead>
<tr>
<th>Temp (°C)</th>
<th>% Water in Feed</th>
<th>% Water in Permeate</th>
<th>Flux (Kg/m².h)</th>
<th>% Water in Feed</th>
<th>% Water in Permeate</th>
<th>Flux (Kg/m².h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>20</td>
<td>68.6</td>
<td>8.7</td>
<td>2.5</td>
<td>0.8</td>
<td>1.7</td>
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<tr>
<td></td>
<td>40</td>
<td>72.3</td>
<td>3.9</td>
<td>2.9</td>
<td>0.8</td>
<td>2.1</td>
</tr>
<tr>
<td></td>
<td>60</td>
<td>73.1</td>
<td>1.8</td>
<td>15.5</td>
<td>4.2</td>
<td>11.3</td>
</tr>
<tr>
<td></td>
<td>80</td>
<td>75.6</td>
<td>0.7</td>
<td>28.7</td>
<td>7.0</td>
<td>21.7</td>
</tr>
<tr>
<td>45</td>
<td>20</td>
<td>76.1</td>
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</tr>
<tr>
<td></td>
<td>40</td>
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<td>3.9</td>
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</tr>
<tr>
<td></td>
<td>60</td>
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</tr>
<tr>
<td></td>
<td>80</td>
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<tr>
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<tr>
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<td>92.1</td>
<td>2.9</td>
<td>36.2</td>
<td>2.8</td>
<td>33.4</td>
</tr>
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</table>

Fig. 3 — Influence of pervaporation on the removal of water of esterification measured in terms of %ethyl acetate formed as a function of esterification time at 90°C.

Monitoring esterification progress by pervaporation

If the water produced during the esterification is removed by pervaporation the reaction equilibrium can be shifted to the forward direction to drive the reaction to completeness. That the pervaporation can effectively remove water of esterification and drive the reaction towards forward direction is evident from a plot of % ethyl acetate formed with esterification time shown in Fig. 3. As seen from Fig. 3 normal esterification has reached equilibrium after 4.8 h at 62% conversion, where water of esterification was not removed by pervaporation. On the contrary when the water was removed by pervaporation a much improvement in conversion (96.5% for PVAMA and 98% for PVACA) was achieved in less than 2.7 h.

Effect of temperature on esterification progress as well as on pervaporation performances was also
investigated for both PVAMA and PVACA membranes and the results are shown in Figs 4 and 5. Running the esterification with simultaneous pervaporation removal of water of esterification at higher temperature (90°C) has shown improvement in the progress of esterification using both the membranes PVAMA and PVACA (Figs 4 and 5) compared to the processes operated at 70 and 80°C. So these results indicate a definite contribution of pervaporation to get higher yield of ethyl acetate. Figures 6 and 7 show the fluxes of permeate components as well as the total flux through the PVAMA and PVACA membranes used during simultaneous esterification and pervaporation. The water flux gradually increased with the progress of esterification and passing through a maximum it decreased. With the progress of esterification there was simultaneous removal of water by pervaporation from the beginning till maximum point so long the water was available as by product. Beyond the maximum point the availability of water gradually decreased due to decrease of esterification rate near completion. So both the total and water flux decrease after maximum. As the membranes are strongly hydrophilic and have affinity towards water the permeation flux of water was much higher than that of acetic acid and ethanol. Again PVACA membrane showed the better water flux than that of PVAMA membrane. The flux result is consistent with the membrane hydrophilicity and molecular sizes of the permeating components.
Conclusion

Poly (vinyl alcohol) membrane crosslinked with maleic acid and citric acid showed good separation capability of water compared to that of acetic acid and ethanol from acetic acid-water and ethanol-water mixtures. Having good water permeation ability of the membranes in pervaporation separation of ethanol-water and acetic acid-water mixtures, the same membrane was used for the pervaporation separation of water by product in ethyl acetate formation. The maximum conversion (98%) to ethyl acetate was obtained using PVACA membrane at a temperature of 90°C. The order of permeation fluxes were: \( J_{\text{water}} > J_{\text{acetic acid}} > J_{\text{alcohol}} > J_{\text{ethyl acetate}} \). Rapid and high conversion to ethyl acetate has shown the utility of membrane pervaporation in ester synthesis with higher efficiency.

Nomenclature

PVA = Poly (vinyl alcohol)  
PVAMA = Poly (vinyl alcohol) crosslinked with maleic acid  
PVACA = Poly (vinyl alcohol) crosslinked with citric acid  
ATR = Attenuated total reflection  
FTIR = Fourier transformed infra red  
\( \alpha_{ij} \) = Separation factor/separation selectivity  
\( J_{\text{water}} \) = Permeation flux of water, kg/m².h  
\( J_{\text{acetic acid}} \) = Permeation flux of acetic acid, kg/m².h  
\( J_{\text{alcohol}} \) = Permeation flux of alcohol, kg/m².h  
\( J_{\text{ethyl acetate}} \) = Permeation flux of ethyl acetate, kg/m².h  
T = Reaction temperature and feed temperature, °C

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