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Design and Application of H-ZSM-5 Zeolite Loaded Hydroxy Propyl Cellulose/ Poly (Vinyl Pyrrolidone) mixed matrix membranes for dehydration of ethanol by Pervaporation

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ABSTRACT

Hydroxypropyl cellulose/poly (vinyl pyrrolidone) (HPC/PVP) mixed matrix membranes (MMMs) filled with different amounts of H-ZSM-5 zeolite (0, 5, 10, and 15 wt.%) were prepared by solution casting method. The prepared membranes were crosslinked with glutaraldehyde and used for pervaporation (PV) dehydration of ethanol at different feed mixtures at 30°C. On increment in zeolite content in the membrane, it is noticed better performance of the MMMs, due to its hydrophilic nature and molecular sieving effect in addition to its favorable interaction with hydrophilic HPC and PVP. The structure, chemical constituents and favorable interactions between polymers, crosslinker, and zeolite in MMMs were investigated by Fourier transform infrared spectroscopy. Thermal stability of the membranes was assessed by differential scanning calorimetry technique. The morphological properties of these membranes were studied by scanning electron microscopy. Sorption studies have been performed to evaluate the extent of interaction and degree of swelling of the membranes with pristine HPC-PVP and H-ZSM-5 zeolite filled blend membranes at different water-ethanol in the feed mixture. The PV experiments results showed that membrane selectivity to water decreased, whereas flux increased with zeolite content in the MMMs. The reason for this can be attributed to the combined effects of molecular adhesion between zeolite and HPC-PVP blend matrix as well as hydrophilicity of the zeolite. The same reason can also be attributed to the observed higher selectivity of the MMMs compared to pristine HPC-PVP blend membrane.

Key words: *Hydroxypropyl cellulose, Poly (vinyl pyrrolidone), H-ZSM-5 zeolite.*

1. INTRODUCTION

Ethanol is an important member of the alcohol family that finds innumerable applications including in paints, coatings, adhesives, cosmetics, pharmaceuticals, and as cleaning agent in semiconductor industries [1]. Ethanol is a renewable "biofuel;" as per the European Union, fuel grade ethanol should contain at least 98.7 wt.% ethanol [2-5]. In many industrial applications, high-purity ethanol (EtOH) is required and is miscible with water in all proportions and form an azeotrope at low water concentrations. For separating this mixture, distillation can be used, but it is effective only to a certain limit (i.e., at a concentration of 80-85 wt.% of alcohol), after which the method becomes costly, especially near the azeotropic point (EtOH 95.6 wt.%) [6]. Therefore, to get high-purity ethanol, the binary mixture has to be concentrated to 80-85 wt.% followed by azeotropic distillation, molecular sieve adsorption, etc. methods [7-11]. Among the other purification techniques, pervaporation (PV) appears to be the method with the most potential to separate liquids as it has a low energy consumption and is easily performed [12], but its success depends on developing a suitable membrane that can selectively separate the component liquid from its mixture [13,14]. Usually, hydrophilic membranes prepared from polymers such as poly (vinyl alcohol), chitosan, sodium alginate, polyacrylonitrile, and polyimide, have been widely employed in alcohol dehydration studies [10,15-17].

In the past decade, efforts to develop durable PV membranes include those of crosslinked polymers, blends, grafts, and nanocomposite membranes [10]. To develop nanocomposite membranes, suitable nanofillers have been added in a polymer matrix [18-20]. Fillers such as clay [21], heteropolyacid

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[22], zeolite [23], mesoporous silica [24], single-walled aluminosilicate nanotubes [25], zeolite immedazolite framework [26], titanate nanotubes [27], carbon nanotubes [28], and graphene [29] have also been employed.

The zeolite-incorporated polymer membranes have received much attention recently in gas and PV separation studies [30,31]. The incorporation of zeolite or porous fillers in the dense membrane can improve the separation performance of the membranes [30-33] due to combined effect of molecular sieving action, selective absorption, and difference in diffusion rates. In addition, zeolites have high mechanical strength, good thermal and chemical stability, and thus, the membranes, when incorporated with these fillers, can be used over the wide range of operating conditions.

The microporous aluminosilicates (zeolites) possess good material properties such as molecule seiving action, selective adsorption, differential diffusion rates, high mechanical strength, thermal stability and hence [34,35], the mixed matrix membranes (MMMs) containing zeolites as fillers in the polymer matrix have been widely explored in PV dehydration [36-38]. In particular, ZSM-5 with MFI (Mobil5) type structures have been used to develop PV membranes [39-41], as the straight channels where ZSM-5 zeolites are elliptical, with a diameter of 0.54 nm [42], would facilitate small molecules transport to achieve the separation.

Among the natural polymers, hydroxypropyl cellulose (HPC) has been widely used by several workers for PV studies because it is an alkyl-substituted hydrophilic cellulose derivative that not only has phase transition behavior in aqueous solution but is also miscible with poly (vinyl pyrrolidone) (PVP). The material is soluble in water as well as in polar organic solvents, but also has many advantages such has excellent film forming properties, biodegradability, and biocompatibility, etc. PVP is another important biopolymer used in PV dehydration studies [34]. To improve PV separation characteristics of water-ethanol mixtures, in this work blending of HPC with PVP is considered to limit the excessive swelling of HPC. Since HPC and PVP polymers are completely miscible in all proportions due to hydrogen bond formation between donor groups of HPC and acceptor groups of PVP, in the blend system, it is thought that selectivity of HPC to water might be enhanced, thus favoring the dehydration of ethanol. In the present study, it is planned to prepare blend membranes of HPC/PVP and incorporated with H-ZSM-5 aiming to obtain high permeability and selectivity simultaneously for PV separation of ethanol-water mixtures.

2. MATERIALS AND EXPERIMENTAL METHODS

2.1. Materials

HPC (MW=1,40,000) was purchased from Sigma-Aldrich Chemicals Company, Milwaukee, WI, USA. Poly (vinyl alcohol) (PVP) having a viscosity average molecular weight 70,000, acetone, hydrochloric acid were purchased from S.D. Fine Chemicals, Mumbai, India. H-ZSM-5 zeolite received as a gift sample from Poornaprajna Institute, Bangalore, India. Ethanol was purchased from Qualigens fine chemicals, Mumbai, India and glutaraldehyde (GA) was purchased from Merck chemicals Mumbai, India. Deionized water having a conductivity of 20 µS/cm was used for the preparation of feed solutions, which was generated in the laboratory itself.

2.2. Membrane Preparation

H-ZSM-5 filled blend membranes of HPC/PVP were prepared by solution casting method. 2 g of PVP was dissolved in 45 mL of distilled water at 90°C, and 2 g of HPC was dissolved in 45 mL of distilled water at room temperature (30°C). These two solutions were mixed thoroughly by constant stirring the mixture for 30 min to form a homogeneous solution. After uniform mixing, the solution was filtered to remove any suspended particles. The 5, 10, and 15 wt.% of H-ZSM-5 particles (with respect to the weight of the polymer) dispersed in 10 ml DI water separately by sonication for 30 min; these two solutions were separately added to the previously prepared blend solution (of 90 ml) and then the whole mixture was kept under stirring for another 24 h. The blend solution was poured onto to a preleveled glass plate in the dust-free atmosphere to cast the membranes with uniform thickness. Membranes thus formed were allowed to dry at an ambient temperature and then peeled off from the glass plate and then crosslinked with GA (2.5 ml) containing 85:10 acetone water mixture in which 2.5 ml HCl was added as an activator and allowed for crosslinking reaction for 2 h. Acetone being a non-solvent prevents the initial dissolution of the membrane and water present in the feed leading to swell the membrane thereby facilitating an easy penetration of GA into the membrane matrix for a sufficient crosslinking. Crosslinking reaction takes place between the -OH groups of PVP/HPC blend, and -CHO groups of GA because of formation of ether linkages by eliminating water molecules. The crosslinked membranes were removed from crosslinking bath and washed repeatedly with distilled water to remove the adhered GA and unreacted molecules and then dried in hot air oven at 50°C to constant weight.

Membranes were designated as M0 when there was pristine membrane and H-ZSM-5-filled membranes were prepared in the same manner by dispersing 5 wt.%, 10 wt.%, and 15 wt.% of H-ZSM-5 and designated as

M1, M2, and M3, respectively. Membrane thickness was measured using a micrometer screw gauge and was found to be around 35-40 µm.

2.3. Swelling Studies

The swelling experiments on the circularly cut membranes were performed at 30°C gravimetrically [43] in 10, 12.5, 15, and 17.5 wt.% water-containing fed mixtures. MMMs of Blend samples with compositions ranging from 10 to 17.5 wt.% water at 30°C±0.5°C in an electronically controlled incubator (WTB Binder, model BD-53, Tuttilgen, Germany) as per procedures reported previously [44]. To do this, dry weight of the circularly cut (dia=2.5 cm) disc-shaped MMMs of blend membranes were stored in a desiccators over anhydrous calcium chloride maintained at 30°C for about 24 h before performing the swelling experiments. This dry weight of the circularly cut (dia=2.5 cm) MMMs of blend membranes was taken using a single-pan Adam digital microbalance (model AFP 210 L) having a sensitivity of ±0.01 mg. The swollen membranes were weighed immediately after careful blotting to remove surface adhered water. The percentage degree of swelling (DS) was calculated as:

$$\%\ \mathrm{DS} = \left(\frac{\mathrm{Ms}}{\mathrm{Md}}\right)\times 100 \tag{1}$$

Where M_S and M_d are the mass of the swollen and dry membranes, respectively.

2.4. PV Experiments

The equipment used to perform the PV experiments remained the same as described earlier Sudhakar *et al*. [45,46]. The PV cell consisted of two bell-shaped B-24 size glass column reducers/couplers clamped together with external padded flanges by means of tie rods to give a vacuum tight arrangement. The top half was used as the feed chamber, and the bottom one worked as the permeate chamber. The membrane was supported by a stainless steel porous plate which was embedded with a mesh of the same material to provide a smooth uniform surface. Teflon gaskets were fixed by means of high vacuum silicone grease on either side of the membrane, and the sandwich was placed between the two glass column couplers and secured tightly. The effective membrane area that was in contact with feed was 20 cm^2. After fixing the membrane, the cell was installed in the manifold and connected to the permeate line by means of a B-24 glass cone which was fixed on one side to a high vacuum glass valve followed by a glass condenser trap which consisted of a small detachable collector. The trap was placed in a Dewar flask containing liquid nitrogen for condensing the permeate vapors. A rotary vacuum pump was used to maintain the permeate side pressure which was measured with an Edward's Mcleod gauge of scale in the range 0.01-10 mmHg. High vacuum rubber tubing was used to connect the various accessories to the experimental manifold. All glass cone-socket joints were fixed with good quality high vacuum grease (Dow Corning, USA).

Initially, the membrane was soaked in the feed solution overnight to attain equilibrium. During the experiments, the membrane upstream side was maintained at atmospheric pressure, and the downstream side pressure was controlled by adjusting the value for vacuum release (vent). The permeate was condensed in the trap for 6-8 h and then collected in a simple bottle for evaluation of its weight to determine the flux and analyzed by gas chromatography to calculate the selectivity. Flux (Ji) was calculated using the Equation (2). The feed was kept in a continuous stirring mode using an overhead stirring motor to minimize the concentration polarization.

$$\mathrm{Ji}=\mathrm{Wi}/\mathrm{At} \tag{2}$$

Here, Wi represents the mass of water in permeate (kg), A is the membrane area (m^2), and t represents the permeation time (h). The selectivity of the H-ZSM-5 zeolite-filled membranes was evaluated by Equation (3). Membrane selectivity, α, is the ratio of permeability coefficient of water to that of ethanol, which is calculated from their respective wt. concentrations in feed and permeate as given below:

$$\alpha=y\ (1-x)/x\ (1-y) \tag{3}$$

Where y is the permeate weight fraction of the faster permeating component (water) and x is its feed weight fraction.

2.5. Measurement of Refractive Index

Refractive index, N^D for sodium–D line was measured using the thermostatically controlled Abbe refractometer (Atago 3T, Japan) with an accuracy of ±0.001. Refractometer was fitted with hollow prism casings through which water was circulated. The experimental Temperature of the prism casing was observed with a digital display (±0.01°C). The instrument directly gives the values of N^D. Permeate composition was determined by measuring refractive index and comparing it with the established graph of refractive index versus mixture composition.

2.6. Fourier Transform Infrared (FTIR) Spectroscopic Studies

FTIR spectra measurements were recorded in the wavelength region of 4000-400 cm^{-1} under an N_2 atmosphere at a scan rate of 25 cm^{-1} using FTIR spectrometer (Bomem MB: 3000, Canada), equipped with attenuated total reflectance. About 2 mg of the sample was grind thoroughly with KBr, and pellets were made under a hydraulic pressure of 600 kg/cm^2.

2.7. Differential Scanning Calorimetry (DSC) Studies

DSC curves of the polymer films were examined using TG instruments (Model: SDT Q600, U.K) in the temperature range of 40-600°C at a heating rate of 10°C/min, with nitrogen flushed at 100 ml/min. The samples were subjected to DSC for both before and after incorporation of zeolite to determine the thermal stability and decomposition characteristics.

2.8. Scanning Electron Micrograph (SEM) Studies

SEM micrographs of surface of the membranes were obtained under high resolution (×300, 5 kv) using JOEL MODEL JSM 840A Japan, SEM, equipped with phoenix energy dispersive. SEM micrographs were taken at Sathyabama University, Chennai.

3. RESULTS AND DISCUSSION

The possible interactions between H-ZSM-5 zeolite and HPC/PVP blend membranes are shown in the Scheme 1.

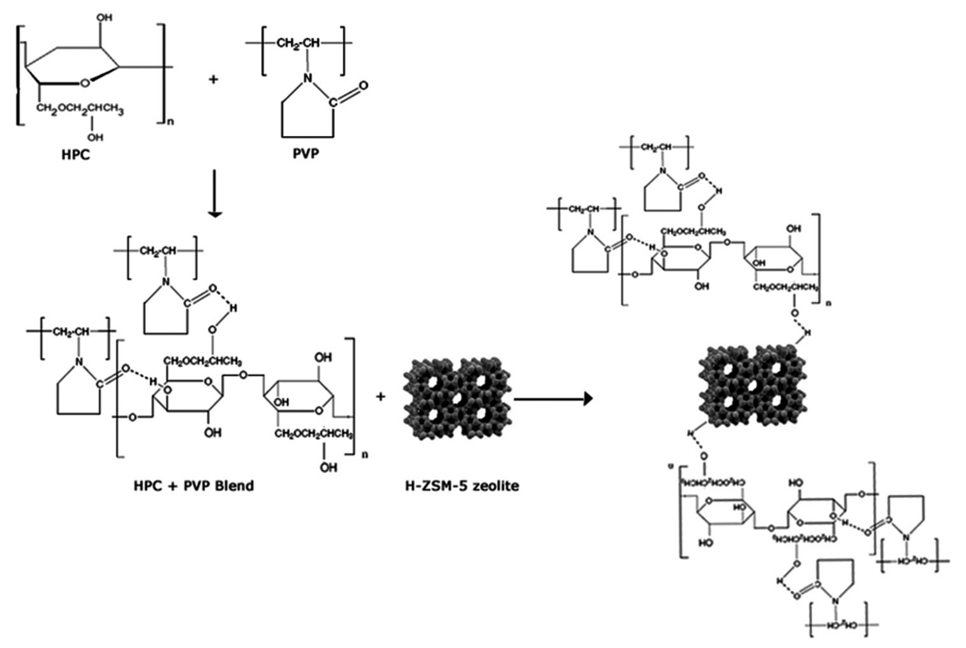
3.1. FTIR Spectroscopy

Figure 1 shows the FTIR spectra of pure H-ZSM-5 (a), pristine HPC/PVP blend membrane (b), and mixed matrix blend membrane of 5wt.% H-ZSM-5 zeolite (SAR-240) loaded membranes(c). From the spectra of Figure 1b, the peak observed at 3410 cm^{-1} corresponding to O-H stretching vibrations of the hydroxyl groups of HPC/PVP blend membrane. Multiple peaks observed at 1000-1600 cm^{-1} region due to the formation of acetal linkage between hydroxyl group of HPC-PVP blend and an aldehydic group of GA. From the Figure 1a and 1c, it is noticed that Al-O vibrations are due to the presence of zeolite [41,42].

The intensity of these bands increased incorporating Zeolite into the HPC-PVP matrix, Figure 1b and Figure 1c new peaks appeared at 1180 cm^{-1} corresponding to Si-O stretching, while two other peaks observed at 816 cm^{-1} and 460 cm^{-1} corresponding to stretching further with the presence of zeolite in the membrane, which has a clear indication of the complete dispersion of zeolite in the crosslinked HPC-PVP membranes.

3.2. DSC

The thermogravimetric analysis of crosslinked unfilled HPC-PVP blend membrane (M0), and H-ZSM-5 zeolite filled HPC-PVP MMMs are determined and discussed here. Figure 2 shows the melting thermograms of crosslinked unfilled HPC-PVP blend membrane (M0) (2a) and 5wt.% (M1) and 10wt.% (M2) H-ZSM-5 zeolite filled (Figure 2b and 2c) respectively. From Figure 2a, it is observed that (M0) has a T_g of 48°C, which is shifted to higher temperatures of 90-110°C for M1 and M2. After addition of H-ZSM-5 zeolite, this may be attributed to intermolecular H-bonding interactions between the blend polymer and zeolite particles. This would decrease the molecular chain mobility of HPC-PVP blend membrane due to the presence of zeolite. From Figure 2a (M0), it can be seen a relatively large and sharp melting endothermic peak at 225°C. On the other hand, weak and broad melting endothermic peaks of HPC-PVP systems in the MMMs (Figure 2b [M1] and Figure 2c [M2]) appeared between 450-475°C. As the content of



Scheme 1: Schematic representation of the interaction between H-ZSM-5 and hydroxy propyl cellulose/poly (vinyl pyrrolidone) blend membrane.

H-ZSM-5 increased, the endothermic curves of HPC-PVP segments became broader, and these peaks shifted to higher temperatures. This increase of melting temperature and peak broadening indicates that the ordered association of the of HPC-PVP chain segments was increased by the presence of H-ZSM-5 zeolite.

3.3. SEM

SEM pictures of unfilled blend membrane of HPC-PVP (50:50) (M0), (a) and 5% H-ZSM-5 filled blend membrane of HPC-PVP (50:50) (M1), (b) were shown in Figure 3. Moreover, from these images, it is observed that the membranes casting were proper as they all dipict nonporous structures. Figure 3a shows the complete miscibilities of the two polymers. Figure 3b membrane indicates that it has a thick and dense top layer almost without any pores confirms a molecular level distribution of H-ZSM-5 particles, such a homogeneous mixing of H-ZSM-5 particles in the bulk of the polymer phase would facilitate higher water transport through the membrane due to the creation of channels that are more favorable to the transport of water molecules than ethanol. These typical SEM images of MMM (M1) suggest smooth surface for this membrane with no phase separation and suggesting a homogeneous and more uniform distribution of H-ZSM-5 particles into MMM of HPC-PVP (M-1) which can be seen in this SEM photograph.

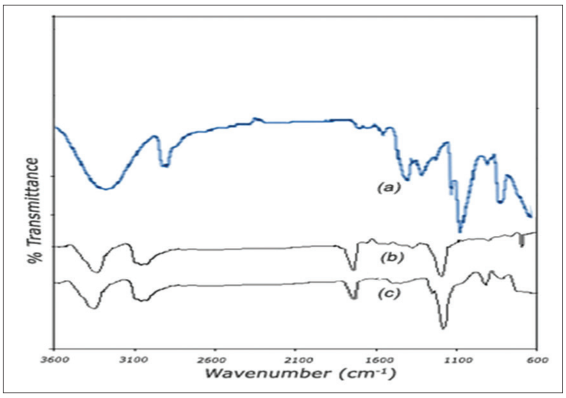


Figure 1: Fourier transform infrared spectroscopy spectra of (a) Pure H-ZSM-5, (b) M0, (c) M1 membranes.

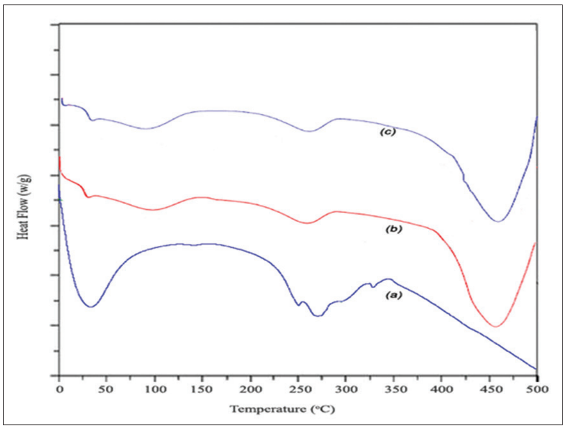


Figure 2: Differential scanning calorimetry thermograms of unfilled hydroxy propyl cellulose/ poly (vinyl pyrrolidone) blend membrane (a), and H-ZSM-5 loaded membranes with 5 wt.% (b), and 10 wt.% (c).

4. MEMBRANE PERFORMANCE

4.1. Swelling Experiments

The degree of swelling obtained from sorption experiments at 30°C for crosslinked pristine blend membrane of HPC-PVP and crosslinked mixed matrix of HPC-PVP blend membranes are measured as a function of wt.% of water in the feed are displayed in Figure 4; and are also shown in Table 1, where Mo 0%wt. (Mo), 5 wt.% (M1), 10 wt.% (M2), and 15wt.% (M3) zeolite filled blend membranes.

In the PV experiment, membrane swelling is an important factor that controls the transport of permeating molecules under the chemical potential gradient. In an effort to study the effects of feed composition and zeolite loading on membrane swelling, the percentage degree of swelling of all the membranes was plotted as a function of the different mass percentage of water in the feed at 30°C, as shown

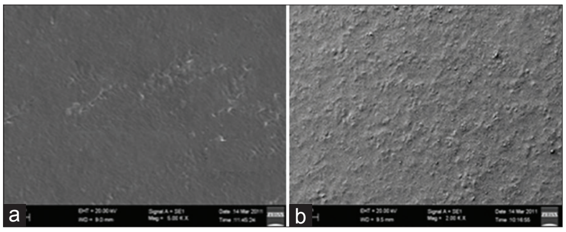


Figure 3: Surface scanning electron micrograph images of unfilled hydroxy propyl cellulose-poly (vinyl pyrrolidone) blend membrane (M0) (a), and H-ZSM-5 filled membrane (M1) (b).

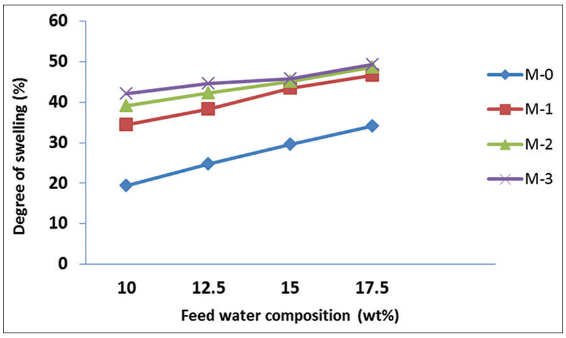


Figure 4: Degree of swelling of prestine hydroxy propyl cellulose/poly (vinyl pyrrolidone (HPC/PVP) blend membrane (M0) and mixed matrix membranes of HPC/PVP blend membranes (M1, M2 and M3).

in Figure 4 and also shown in Table 1. It is observed that the degree of swelling increased exponentially for all the membranes with increasing mass percentage of water in the feed. This is attributed to strong interactions between the hydroxyl and COO^- groups of HPC and C=O groups of PVP with the water molecules. As the water concentration increased in the feed, the interaction with water becomes more predominant, given that water causes a greater degree of swelling than that of ethanol with the membrane. On the other hand, when the polymer matrices are filled with highly symmetrical zeolite particles, then it is likely that the pores of the membranes might have been occupied by the zeolite particles. This leads to an increased degree of swelling over that of the pure HPC-PVP membranes, as observed in Figure 4. However, the effect is significantly increased from membrane 5 wt.% to 15 wt.% H-ZSM-5 zeolite filled HPC-PVP membranes with increasing zeolite content. This is mainly because of the hydrophilic nature of the incorporated H-ZSM-5 zeolite in the membrane matrix.

The increased sorption is due to the presence of zeolite particles that possess three-dimensional channel network with an asymmetrical aperture, which helps to increase water uptake capacity of the membrane compared to the organic components of the feed. This also would result in increased separation factor for filled matrix membranes compared to the pristine HPC-PVP membrane.

Table 1: Percentage swelling data of Ethanol and water mixtures at 30°C for different blend membranes.

% of water in the feed	% of swelling			
	M0	M1	M2	M3
10	19.36	34.54	39.22	42.25
12.5	24.78	38.26	42.35	44.76
15	29.56	43.45	45.12	45.87
17.5	34.56	46.73	48.68	49.36

In general degree of swelling depends on the extent of interaction of water molecules with the membrane material and hence, higher swelling would mean higher thermodynamic interactions. However, the extent of interaction depends on the nature of the organic component of the feed mixture. For water-Ethanol feeds, the higher polar ethanol molecule allows lesser number of water molecules to penetrate through the HPC-PVP membrane. From the results, it is observed that zeolite filled membranes swell more than the pristine HPC-PVP. This indicates that more swelling will be there with the more amount of zeolite which in turn indicates hydrophilic nature of the zeolite. Since the polymer gets plasticized in the presence of large amount of water, this, in turn, allows the membrane to absorb more of water molecules in the presence of hydrophilic clay fillers, leading to still higher level interactions with water.

5. PV STUDIES

5.1. Membrane Performance on Flux and Selectivity

PV performance of pristine HPC-PVP, 5 wt.% and 10 wt.% H-ZSM-5 zeolite MMMs have been studied by calculating flux and selectivity, for water+ethanol mixture at 30°C and the values are presented in Table 2. The MMMs exhibited better separation characteristics than the pristine membrane. Figures 5 and 6 show the variation of flux and selectivity with mass percentage of water in the feed for different wt.% of zeolite particles into HPC-PVP matrix, respectively. Pristine HPC-PVP membrane exhibited the lowest selectivity of 32 with a flux of 0.1236 kg/m^2 h at 10 wt.% water-

Table 2: PV data for feed mixture of water-ethanol at 30°C.

Wt.% of water in feed	Wt.% of water in permeate	Flux (J) Kg/m^2 h	Selectivity (α)
Pristine (HPC/PVP) (M0)			
10.0	78.26	0.1236	32
12.5	80.24	0.1566	28
15.0	81.56	0.1782	25
HPC/PVP (M1)			
10.0	98.56	0.2068	616
12.5	97.48	0.2274	270
15.0	97.08	0.2543	188
HPC/PVP (M2)			
10.0	98.82	0.2458	753
12.5	97.56	0.2674	279
15.0	97.12	0.2842	191

HPC: Hydroxy propyl cellulose, PVP: Poly (vinyl pyrrolidone), PV: Pervaporation

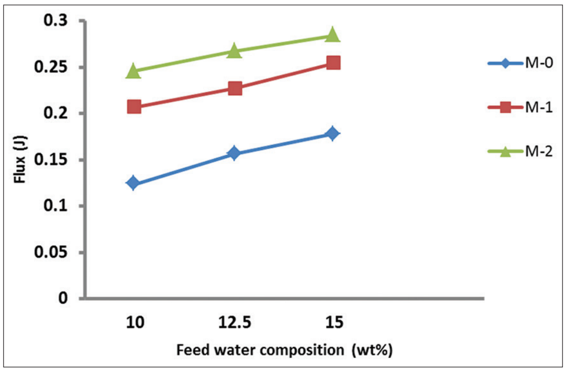


Figure 5: Flux versus wt.% of water in feed mixture for pristine hydroxy propyl cellulose/poly (vinyl pyrrolidone) (HPC/PVP) blend membrane (M0) and mixed matrix membranes of HPC/PVP-1(M1), HPC/PVP (M2).

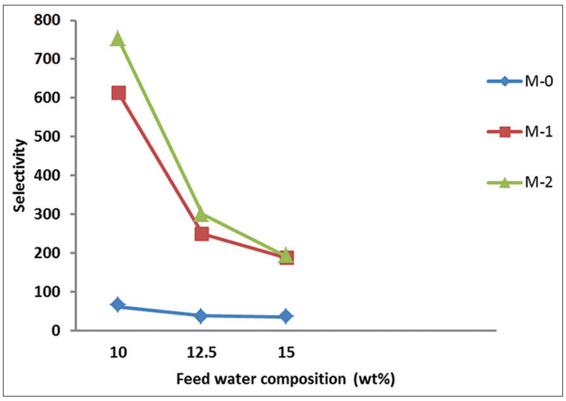


Figure 6: Selectivity versus wt.% of water in feed mixture for pristine hydroxy propyl cellulose/Poly (vinyl pyrrolidone) (HPC/PVP) blend membrane and mixed matrix membranes of HPC/PVP-1(M1), HPC/PVP (M2).

containing feed mixture of water. However, the flux and selectivity values of HPC-PVP membrane increased after incorporation of zeolite particles. For 5 wt.% ZSM-5 zeolite filled HPC-PVP membrane, the selectivity has increased to 616, while that for 10 wt.% H-ZSM-5 zeolite HPC-PVP membrane; it was increased to 753 at 10 wt.% of water in the feed mixture of water; whereas the values of fluxes were 0.2068 kg/m^2 h and 0.2458 kg/m^2 h, for 5 wt.% and 10 wt.% H-ZSM-5 zeolite filled HPC-PVP MMMs (M2), respectively.

Zeolite-polymer MMMs are quite useful in PV dehydration studies because of incorporation of zeolites they swell more due to the absorption nature of the fillers. Expectedly, zeolite, through the molecular sieving effect, would also allow selective sorption and diffusion of liquid molecules as a result of decrease in the mobility of the less permeable component of the feed mixture [31]. Besides, the molecular sieving effect, zeolite hydrophilic interactions would also contribute to the facile transport. When the zeolite particles are added to the HPC-PVP matrix, the MMMs becomes hydrophilic, and thus, the organic components of the feed are retained back at the feed side of the membrane. At 10 wt.% incorporation of zeolite into HPC-PVP matrix, membrane swelling capacity has increased. Membranes prepared by adding more than 10 wt.% of particles were found to be more flexible than those containing lower amounts of particles and hence, these were useful in the PV study.

5.2. Effect of Feed Water Composition

In the present research, it is demonstrated that by incorporating zeolite particles, one can improve the solvent stability of HPC-PVP membrane, thereby improving the membrane performance. As displayed in Figures 5 and 6, flux and selectivity values of HPC-PVP MMMs are higher than those of pristine HPC-PVP. For HPC-PVP MMMs, flux values increased with increasing feed water composition from 10 to 15 wt.%. As the water composition of feed mixture increases, the selectivity decreases. As seen from Figure 6. HPC-PVP blend membrane swells to a greater extent at higher feed water composition, which induces the plasticization to membranes. Thus, sorption results are a good indicator of PV performance of the membranes. At 15 wt.% of water in the feed mixture containing water-ethanol, selectivity for pristine NaAlg membrane is quite small, i.e. 25 with a flux of 0. 1782 kg/m^2 h, whereas 5 wt.% of H-ZSM-5 zeolite and 10 wt.% H-ZSM-5 zeolite mixed membranes showed selectivities of 616 and 753 respectively with the flux values of 0.2068 and 0.2543 kg/m^2 (Figures 5 and 6, Table 2).

6. CONCLUSIONS

The present study deals with the development of novel type of MMMs from the blend polymers of HPC and poly (vinyl pyrrolidone) (PVP) incorporated with hydrophilic H-ZSM-5 particles for the use in effective ethanol dehydration. The boost in PV performance of the MMMs is attributed to hydrophilic-hydrophilic interactions between the filler nanoparticles and water. At higher loadings of H-ZSM-5 nanoparticles, aggregation effect may be responsible to reduce water selectivity. The flux and permeate values increased with increasing feed water composition. Sorption data suggest that membrane selectivity is mainly governed by sorption selectivity. With increasing feed water composition the membrane performance exhibited a reduction in selectivity and improvement in flux due to increased swelling. On the whole, the PV separation MMMs of HPC/PVP blend membranes (M1&M2) are better than pristine HPC/PVP (M0) membrane. This work suggests higher water-selective nature for the developed MMMs.

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