

Pervaporation Separation Of Water-Isopropanol Mixture Using MFI-24Q Zeolite Incorporated Blend (Naalg And HPC) Membranes

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Abstract: Using solution casting technique, MFI-24Q Zeolite-incorporated (Sodium alginate and Hydroxy propyl cellulose) blend membranes were prepared and subjected to characterized and used for the pervaporation separation of water-isopropanol mixtures. Fourier transform infrared spectroscopy technique was used to analyze the possible chemical reactions between NaAlg, HPC, zeolite, and glutaraldehyde. TGA studies revealed the stability of the zeolite incorporated membrane and scanning electron microscopy results used to analyze the surface morphology of the membranes. X-Ray diffraction analysis showed the broadening and sharpening of the peaks at different NaAlg concentration with HPC. This indicated changes in the crystallinity to amorphous ratio which in turn showed the miscibility of these two polymers in the blend membrane. The effects of Zeolite loading and feed composition on the pervaporation performance of the membranes were analyzed. Both flux and selectivity increased simultaneously with an increase of zeolite content in the polymer matrix. This was explained on the basis of enhancement of hydrophilicity, selective adsorption and molecular sieving action including reduction of pore size of the membrane matrix. The membrane containing 15 mass % of zeolite shows the highest separation selectivity of 991 with a flux of 0.67×10^{-2} kg/m² h for 10 mass % of water in the feed mixture at 30°C. Increase in water selectivity of the membrane was explained due to a reduction in free volume by increasing zeolite content of the membrane. Separation selectivity and permeation flux data are dependent on water composition of the feed mixture. The hindrance of water permeation at higher composition of water in the feed mixture was explained due to the formation of clusters of water molecules.

Keywords: separation techniques; Membranes; Zeolite; selectivity, Activation energy.

1 INTRODUCTION

Pervaporation (PV) allows isolation of valuable compounds from liquid mixtures that are difficult to separate by distillation, extraction and absorption. Pervaporation is especially advantageous for separation of azeotropes, close boiling mixtures and thermally sensitive compounds and organic compounds present in trace concentrations. Polymeric membranes have limits in their performances for various applications due to tradeoff effect between permeability and selectivity, which means that membranes of more permeable nature are generally less selective and vice versa. To overcome these problems, many researchers attempted to use inorganic materials as a modifier such as zeolite, silica, and carbon molecular sieve to improve the performances of the polymer membranes.

Hydrophilic zeolite membranes are capable of removing water from its mixtures with organic compounds through pervaporation. Zeolite membranes are thus well suited for separating liquid-phase mixtures by pervaporation and the first commercial application of zeolite membranes has been for dehydration of organics. Because of the large number of zeolites that can be prepared, such membranes can be used to remove water from organics or even extract organics from water. The first reported zeolite membranes were prepared by Suzuki, [1] and now more and more number of different zeolite structures have been used as membranes [2, 3]. MFI-24Q zeolite is an inorganic crystalline structure with uniform, molecular-sized pores and high mechanical, thermal and chemical stability and has been used in membrane applications [4-8]. Zeolite incorporation often increases both pervaporation flux and selectivity [9] or increases separation factor with little or no decrease in flux [10-12]. Polymeric systems that exhibited improved separation factor and flux upon zeolite incorporation include alcohol dehydration with (NaAlg + HPC) blend membranes filled with MFI-24 zeolite [13-14]. Sodium alginate (NaAlg), a naturally occurring polysaccharide extracted from seaweed is a linear copolymer of β - (1-4) - linked D-mannuronic acid and α -(1-4) linked L-guluronic acid units. It is found to have an excellent performance as a membrane material for pervaporation-based dehydration of organic solvents[15]. Hydroxypropylcellulose, a nonionic water-soluble polymer, has been widely used in pharmaceutical applications [16, 17] and it is also used as a binder and as a thickening agent in many topical products [18]. Due to its excellent film forming property and hydrophilic nature, most studies mainly focused on modified HPC [19], HPC hydrogel [20], cross linked HPC nanoparticles [21] and HPC blends with PVA, PAA, PAANA, CH, PVME, PVP, PAN etc. based on hydrogen bonding.[20, 22-24]. However, studies on the miscibility of HPC with NaAlg are very rare. In this investigation, a solution blending system of polymers

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containing hydrogen bonding groups [23, 25] were used with HPC and NaAlg. In the present study, blending of NaAlg with HPC has been explored for pervaporation based separation, because both the polymers contain H-bonding groups. NaAlg-HPC blend solution was incorporated with a hydrophilic MFI-24Q Zeolite in different ratios. The structure, morphology, thermal properties and mechanical properties of these membranes were systematically investigated. Effects of MFI-24Q zeolite content on the sorption and diffusion properties of the mixed matrix membranes were discussed. Isopropanol (IPA) is one of the most important solvents used on a large scale in the chemical industry based on oils, gums, waxes and cosmetics as well as in pharmaceutical laboratories. Further, it is also used in semiconductor and liquid crystal display industries as water-removing agent [26,27]. Isopropanol is also more effectively used in medical applications. Isopropanol and water form an azeotropic mixture at 87.5 mass % of alcohol [28] and hence, the separation of these mixtures by conventional methods such as solvent extraction, rotavapor or distillation could prove uneconomical for obtaining absolute IPS. Several membrane materials have been modified recently for the selective separation of water from aqueous mixture of isopropanol [29]. However, the membranes employed in such separation studies often yield compromised values of flux and selectivity due to trade-off phenomenon existing between the flux separation factor in PV process. This study aims to improve the separation factor without any reduction in flux through the use of mixed matrix membranes based zeolite loaded NaAlg-HPC blends.

2 Experimental

2.1 Materials

Sodium alginate (NaAlg) having a viscosity average M.W. \approx 500,000 was purchased from S.D. Fine chemicals, Mumbai, India. Hydroxypropylcellulose (HPC) with M.W. \approx 15,000 was purchased from Aldrich Chemical Company, Milwaukee, WI, USA. Zeolite MFI-24 Q (Nano) and Isopropanol were purchased from Qualigens fine chemicals, Mumbai, India. Glutaraldehyde was purchased from Merck Specialties chemicals, Mumbai, India. Acetone & Hydrochloric acid were purchased from S.D. fine chemicals, Mumbai, India. Demineralized water having a conductivity of 0.02 S/cm, which was used for the preparation of feed solution, was generated in the laboratory itself.

2.2 Preparation of (NaAlg + HPC) Blend membrane

3.04 gms of Sodium alginate and 0.76 gm of HPC was dissolved in 80ml of H₂O with constant stirring (Remi Equipment Model 2 MLH, Mumbai, India) for about 24h at room temperature. Separately 0.2, 0.4 and 0.6 gm of Zeolite particles were weighed and dispersed in 20ml of water by sonicating for 2Hrs and added to the prepared blend (NaAlg + HPC) solution and stirred for a further period of 24 Hrs. The solution was poured on a dust free glass plate to cast membranes of uniform thickness. Dried membranes were peeled off from the glass plate and immersed in a cross linking bath containing (30:70) water acetone mixture along with 2.5 ml of GA (Glutaraldehyde) and 2.5 ml of concentrated HCl. After placing the membranes in this solution for 12 – 14 hrs they are washed

repeatedly with deionized water and dried in an oven at 60°C. Pristine NaAlg membrane was also prepared in the same manner except that clay filler particles were not added. Membranes thickness as measured by a micrometer screw gauge was around $50 \pm 1.0 \mu\text{m}$. Hybrid composite membranes for this study were slightly cloudy white and semi transparent.

2.3 Swelling measurements

The degree of swelling of Zeolite-incorporated membranes was determined in different compositions of water and isopropanol mixtures for 24 h at 30°C using an electronically controlled oven (WTB Binder, Germany). The masses of the dry membranes were first determined. The dry membranes were equilibrated by soaking in different composition of the mixture in a sealed vessel, at 30°C for 24h and then the swollen membranes were weighed immediately after careful blotting the moisture with a filter paper and weighing on a single pan digital microbalance (Model, AFP210L) having a sensitivity of $\pm 0.01\text{mg}$. The % degree of swelling (DS) was calculated as:

$$(1) \quad DS (\%) = \left(\frac{W_a - W_o}{W_o} \right) \times 100$$

Where W_a and W_o are the mass of the swollen and dry membranes, respectively.

2.4 Pervaporation (PV) procedure:

Pervaporation experiments have been carried out using an indigenously designed apparatus reported in the previous articles [30, 31]. The effective membrane area in the PV cell is 34.23 cm² and the capacity of the PV cell is about 250 cm³. The vacuum in the downstream side of the apparatus was maintained ($1.333224 \times 10^3 \text{Pa}/10\text{Torr}$) by a two-stage vacuum pump (Toshniwal, Mumbai, India). The test membrane was allowed to equilibrate for about 2h while in contact with the feed mixture before performing the PV experiment. The procedure used in pervaporation (PV) experiments has been described by Chowdoji Rao et al. [32]. After the steady state was attained, the permeate was collected in trap immersed in liquid nitrogen jar on the downstream side at a fixed time of intervals. The flux was calculated by weighing the permeate on a digital microbalance (Model AFP 210L: accuracy 10^{-4}g) Membrane performance in PV experiments was studied by calculating the total flux (J_p), separation factor (α). These were calculated, respectively, using the following equations.

$$J_p = W_p / At \quad (2)$$

Here W_p represents the mass of water in permeate (kg), A is the membrane area (m²) and t represents the permeation time (h),

$$\alpha = \left(\frac{Y_A}{1 - Y_A} \right) \left(\frac{1 - X_A}{X_A} \right) \quad (3)$$

Where X_A is Wt. % of water in the feed and Y_A is Wt. % of water in the permeate. At least three independent measurements of flux and selectivity were made under the

same conditions of temperature and feed composition to confirm the steady-state pervaporation.

2.5 Measurement of refractive index (RI):

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. Temperature of the prism casing was observed with a display ($\pm 0.01^\circ\text{C}$). The instrument was provided with two prisms placed one above and the other in front of the telescope. Upon inserting a drop of test liquid using a hypodermal syringe, the incident ray forms a line of demarcation between light and dark portions of the field, when viewed with a telescope, which moves with scale. 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 liquid mixture composition.

2.6 Characterization Techniques

2.6.1 Fourier Transform Infrared spectroscopy (FTIR)

FTIR spectral measurements were performed using Bomem MB-3000 (make: Canada) spectrophotometer equipped with KBr disc method. Each sample was finely grounded with KBr to prepare pellets under a hydraulic pressure of 400kg and spectra were recorded between 4000 and 400 cm^{-1} .

2.6.2 Differential Scanning Calorimetric (DSC)

DSC thermo grams of polymer blend membranes, zeolite membranes were recorded using Differential scanning calorimeter (Model – SDT Q600, USA). Initially, the moisture was removed by heating the Samples and then, thermo grams were recorded from 30 to 600 $^\circ\text{C}$ at the heating rate of 10 $^\circ\text{C}/\text{min}$ under nitrogen atmosphere and at a flowrate of 30mL/min. The Sample pan was conditioned in the instrument before running the experiment.

2.6.3 Scanning Electron Microscopy (SEM)

SEM micrographs of the membranes were obtained under high resolution (Mag 300x5kv) Using Leo Model Leica-stereoscan440, scanning electron microscope (SEM), equipped with phoenixenergy dispersive analysis. SEM micrographs were taken at National Chemical Laboratory, Pune.(Courtesy of prashant K Mane, Scientist, Organic Chemistry Division NCL.Lab).

2.6.4 X-ray diffraction (XRD)

A Siemens D 5000 (Germany) powder X-ray diffractometer was used to study the solid –state morphology of the MMMs of SA / HPC blend membranes. The X-rays of 1.5406 Å wavelengths were generated by a Cu K α radiation source. The angle of diffraction (2θ) was varied from 0 $^\circ$ to 65 $^\circ$ to identify any changes in crystal morphology and intermolecular distances between inter-segmental chains of the polymer. The XRD spectra were obtained from university of Hyderabad.

Methods:

Scheme 1 represents the polymers used in the study and also shows the structures of blend (NaAlg-HPC)

incorporated zeolite membrane cross linked with glutaraldehyde, where the –CHO groups of glutaraldehyde, react with the hydroxyl groups of blend membrane incorporated MFI-24Q zeolite resulting in the formation of covalent bond. This can also be confirmed by FTIR Studies. It was noticed that both the homo-polymers and the blend membrane incorporated zeolite were optically clear to the naked eye. No separation into two layers or any precipitation was noticed when allowed to stand for one month at room temperature.



Scheme 1. Structural representation NaAlg/HPC blends membrane crosslinked with glutaraldehyde (GA).

3. Results and Discussion

3.1. Membrane characterization techniques.

3.1.1 FTIR studies

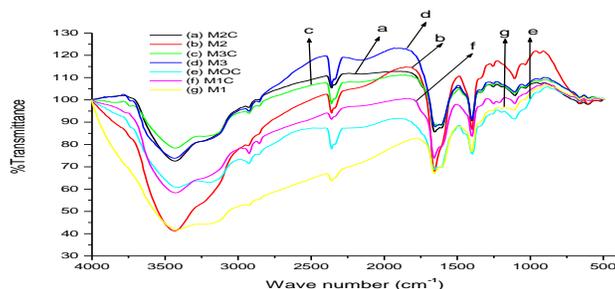


Figure : 1 FTIR Spectra of cross linked and uncross linked blend membranes With and without zeolite : (MoC) 0 mass%; (M1 & M1C) 5 mass%; (M2 & M2C) 10 mass%; (M3 & M3C) 15 mass%.

FTIR spectra of the cross linked and uncrosslinked blend membranes of NaAlg & HPC and those of different amounts of MFI-24 zeolite loaded membranes are shown in Figure:1. A characteristic peak at 3440 cm^{-1} in all the membranes correspond to O-H stretching vibrations of NaAlg, HPC and blend membranes [33] is observed, on the other hand, peak at 2931 cm^{-1} was observed and the related peak at 2360 cm^{-1}

¹ corresponds to C-H stretching vibrations. The peaks at 1658-1604 cm^{-1} are attributed to C=O Stretching vibrations [34]. And the multiple bands appearing around 1396 and 1033 cm^{-1} are assigned to the stretching vibrations due to the presence of zeolite. The intensity of these bands increased further with increasing amount of Zeolite in the membranes, which ascertains the complete dispersion of Zeolite in the cross linked blend membranes. The cross linking between NaAlg & HPC through glutaraldehyde also confirmed the presence of ether linkage which is observed in these figure.

3.1.2. X-ray diffraction studies

The XRD patterns of all the membranes with and without loaded zeolite are presented in Figure.2. The NaAlg / HPC blend membrane without zeolite (M0) exhibited two broad peaks around 10° and 20° in a diffraction pattern (M0), and is due to the existences of amorphous and crystallize regions, respectively. From the pattern, it is observed that after incorporating 5, 10, 15 mass % of zeolite membranes (M1 M2, M3) show the characteristic peaks of both the Zeolite and blend membrane. However, when the Zeolite content increased to 15 mass% (M3), almost no characteristic peak of blend membrane was noticed in the XRD pattern, indicating that blend membrane no longer retains its crystallinity. On the contrary, the resulting membranes tend to have a more rigid structure owing to the restriction of polymer chain mobility with increasing the Zeolite loading in membrane matrix.

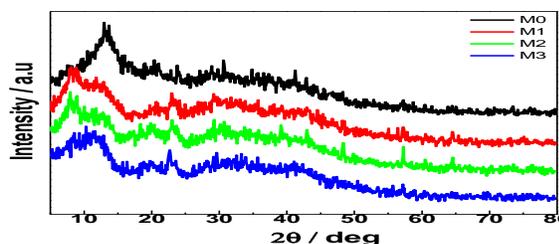


Figure:2 X-ray diffraction patterns of the NaAlg/HPC blend membranes with and without loaded zeolites [(M0) NaAlg-HPC-0,(M1)NaAlg-HPC-5,(M2) NaAlg-HPC-10, (M3) NaAlg-HPC-15].**3.1.3. Scanning electron microscopy (SEM)**

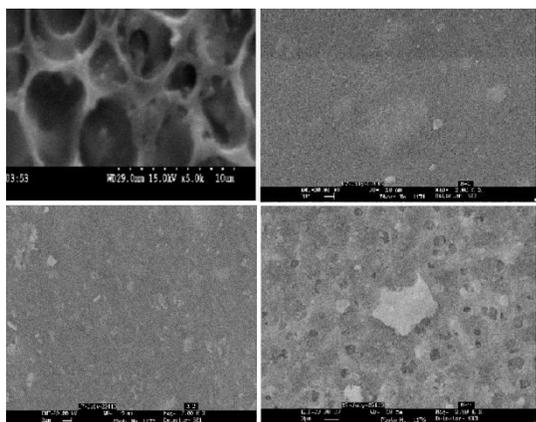


Figure :.3. SEM images of NaAlg/ HPC blend membranes with and without loaded zeolites [(M0) NaAlg-HPC-0; (M1) NaAlg-HPC-5; (M2) NaAlg-HPC-10; (M3) NaAlg-HPC-15.]

Figure.3.1.3 represents the SEM images of surface morphology with and without zeolite loaded NaAlg-HPC blend membranes. The SEM images indicate that the zeolite distribution increased from membrane M1 to M3 with increasing zeolite loading. The zeolite was distributed evenly throughout the membranes, [35] but when the zeolite content reaches 15% wt of the zeolite aggregate at the interface of the membrane that could be disadvantageous for Pervaporation performance. Furthermore, the images clearly show that the zeolite particles are embedded in the membrane with no voids around them (up to 15% zeolite content in the membrane). This ensured the possible defects up to 15% zeolite.

3.1.4. Differential scanning calorimetry: DSC thermograms of pristine blend membrane (M0) and zeolite filled membranes (M1, M2 and M3) are shown in figure: 4. The DSC curve for pristine M0 (a) membrane shows an endotherm at 180°C , which corresponds to its melting point. The melting endothermic peak has shifted to higher temperatures, i.e 200°C (b) and 210°C (c), for M1 and M2 membranes respectively. However, increase in T_m for MFI-24Q zeolite filled blend membrane is due to the increase in the amount of MFI-24Q zeolite filler particles that are inorganic in nature. It is noticed that as the MFI-24Q zeolite particles have higher melting temperature, T_m of MFI-24Q zeolite filled blend membrane matrix also increases.

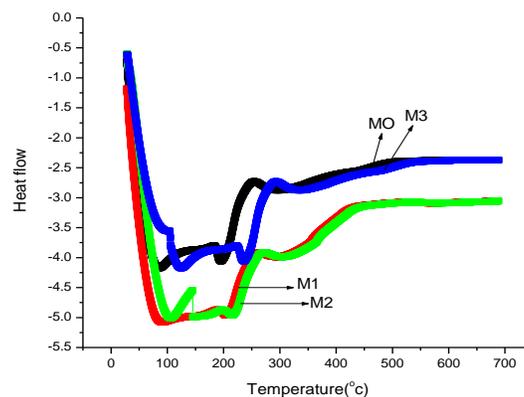


Figure: 4. DSC thermograms of M0 (a), M1 (b), M2(c), M3 (d) Membranes

3.1.5. Thermo gravimetric analysis (TGA)

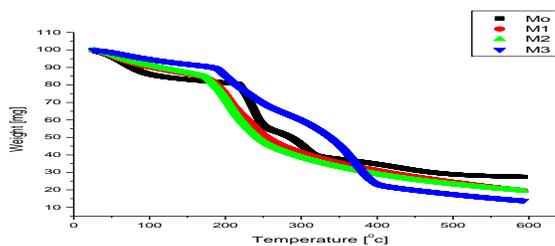


Figure :.5. TGA analysis of (a) M-0; (b) M-1; (c) M-2 and (d) M-3.

From Fig: 5. It is noticed that two weight loss stages in the temperature range between 200 and 350°C followed by the final decomposition of the M-0 membrane that began around 450°C. Weight loss in the first stage could be attributed to splitting of the main polymer chain before its final decomposition. After incorporating MFI-24Q Zeolite; the membranes (M-1 to M-3) exhibited two weight loss stages in the range of 280-450°C and 520-570°C followed by a final decomposition. An increase in thermal stability is observed with increase in zeolite content in the filled blend membranes.

3.1.6. Swelling Results:-

It is proved [36, 37] that the polymer swelling in aggressive media depends on the extent of crosslinking, morphology of the polymer, and the free volume available within the polymer matrix. The transport of permeate molecules is controlled by membrane swelling in the PV experiments. When the polymer matrices are filled with zeolite particles the pores of membranes show different flow of permeation. Because, in the present investigation, hydrophobic property is predominant in the type of zeolite used, it hinders the chain mobility due to loss in free volume. Therefore, both solubility and diffusivity of the permeant molecules through the zeolite-filled membrane might have decreased during the PV process with increased zeolite loading. In the present study the results of % degree of swelling at 30° C for plane blend membrane and zeolite incorporated blend membranes were measured as a function of mass % of water in water + Isopropanol feed mixtures and the results are presented in **Table.1**. These variations are displayed in Fig.:6, which indicates that swelling kinetics depend on the mutual diffusion of solvent molecules in response to the polymer segmental relaxation [38]. Fig.:6. also shows Degree of swelling is higher in case of nascent NaAlg/HPC membranes compared to zeolite incorporated membranes. It is also observed that as the zeolite concentration increases the degree of swelling decreases. The lowest swelling observed for MFI-24Q zeolite is in accordance with the free volume results as with increasing filler loading, the free volume decreases and swelling decreases.

Table: 1 Percentage swelling data of IPA and water mixtures at 30°C

% of water in the feed	% of Swelling			
	M0	M1	M2	M3
10.0	17.02	10.00	10.00	06.66
12.5	50.00	38.46	36.36	17.94
15.0	58.33	53.00	41.66	23.07
17.5	72.15	70.50	62.50	46.15

The higher swelling in zeolite filled membranes compared to pristine membrane is due to the result of performance interaction of water molecules which would accommodate higher amount of water than Isopropanol. This could be possibly due to: (i) Vaporization on the permeate side, (ii) Strong adsorptive hydrophilic interaction of water molecules onto MFI-24Q zeolite particles and (iii) Surface diffusion from cage to cage. Physical adsorption involves both vanderwaals type dispersive-repulsive and electrostatic interactions due to polarization, as well as dipole and

quadrupole type interactions. The complimentary effects of molecular sieve on water transport would thus improve the membrane performance. It is worth mentioning that the varying effects of flux and selectivity are attributed to the amount of ZSM-5zeolite filler present in the NaAlg/HPC matrix.

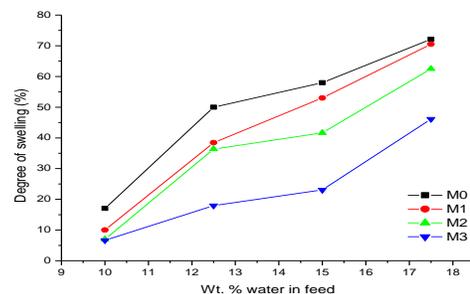


Figure: 6. Degree of swelling Vs percentage of water in different blend membranes M0 (NaAlg-HPC-0) (M1) (NaAlg-HPC-5) (M2) (NaAlg-HPC-10) (M3) (NaAlg-HPC-15).

3.1.7. Effect of MFI-24Q zeolite content on Pervaporation properties.

Table: 2. Pervaporation data of IPA and water mixtures at 30°C and at 1 mmHg permeate pressure

Feed compositions		Permeate compositions(wt %)		Selectivity (α)	Flux (kg /m ² h)	PSI
Water (x)	IPA (1-x)	Water (y)	IPA (1-y)			
M-0						
10	90	97.27	2.73	320	0.0293	9.346
20	80	97.5	2.5	156	0.1173	18.181
30	70	95.5	4.5	49	0.1309	6.283
40	60	90.2	9.8	13	0.1806	2.167
M-1						
10	90	97.43	2.57	341	0.1094	37.19
20	80	97.02	2.98	291	0.1458	18.80
30	70	96.89	3.11	145	0.3959	28.50
40	60	95.44	4.56	68.9	0.4688	11.57
M-2						
10	90	98.82	1.18	753	0.5529	415.78
20	80	96.84	3.16	122	0.5562	67.30
30	70	96.62	3.38	66.7	0.5612	36.87
40	60	95.99	4.01	35.9	0.7545	26.33
M-3						
10	90	99.1	0.9	991	0.670	663.30
20	80	98.64	1.36	291	0.775	224.75
30	70	98.42	1.58	145	0.857	123.40
40	60	97.87	2.13	68.9	0.864	58.66

Zeolites are known to have high affinity for water. As a result, there has been considerable work focused on trying to incorporate zeolites as the active component in a membrane and zeolite incorporated membranes display very high flux and selectivity with IPA-water mixtures. Membrane performance was studied through flux and selectivity data generated at 30°C and presented in **Table:2** PV performance of the cross linked NaAlg-HPC blend membrane and MFI-24 Q zeolite incorporated NaAlg/HPC blend membranes were investigated by varying feed compositions from 10 to 40 wt. % of water (Table.2). The effects of the feed water content on the PV performance of with and without zeolite incorporated (M-0) membranes (M-1, M-2 and M-3) are shown in **Figure: 7** The flux increased and the selectivity decreased with increasing feed water content, which is attributed to the hydrophilicity of the membranes. The increasing water content makes the membranes become more swollen; hence, this increases flux and decreases the selectivity. From **Figure: 7** It can also be noticed that when zeolite content is 15 wt%, the flux of the M-3 membrane is higher than that of the M-0 membrane, and the selectivity of the M-3 and M-0 membranes almost slightly increased. Therefore, the flux for the M-3 membrane was larger than that of the M-0 membrane and the selectivity of the M-3 and M-0 membranes remained almost constant. The effects of feed water content on the PV performance of the M-1 and M-2 membranes are similar to that of the M-3 membranes Figure.3.1.7 also indicates that there is very high difference between the M-0 and M-3 membranes for their PV performance.

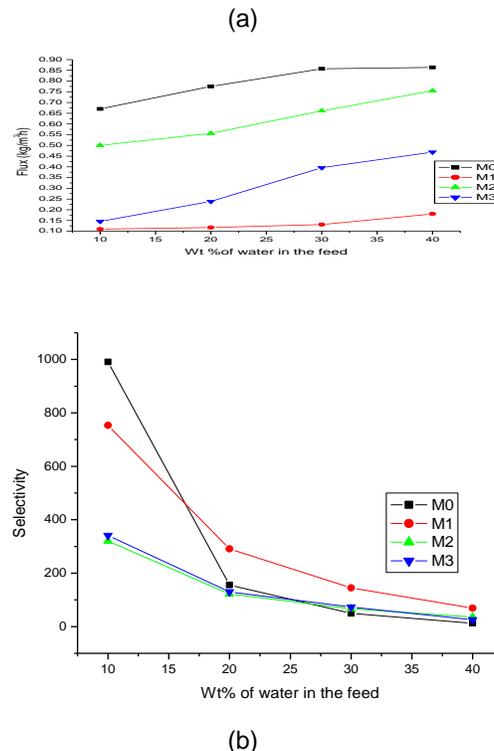


Figure : 7. (a) Effect of wt% of water in the feed on flux for different formulations. (b) Effect of wt% of water in the feed on selectivity for different formulations.

3.1.8.: Effect of feed water composition:

The feed concentration is one of the most important operating factors of a membrane separation process as shown in **Fig.: 8**. Table 2 gives the pervaporation results of MFI-24Q zeolite filled NaAlg/HPC membrane for water-isopropanol mixtures at 30°C. Permeation flux increases with increasing water content of the feed, which is caused due to higher concentration of water molecules within the membrane. At the same time, wt % of water in permeate line and selectivity values decreased for all the water-isopropanol mixtures of this study. There is a two-fold increase in the permeation flux for water-isopropanol mixtures because of the hydrophilic nature of the zeolite used and its interaction with NaAlg/HPC matrix. For all the feed mixtures, flux values of MFI-24Q zeolite-loaded NaAlg/HPC membranes are higher than that of the pristine NaAlg/HPC membrane. The increase in permeation flux for water-isopropanol mixture is purely due to an increase in thermodynamic interactions between water and isopropanol. Membrane performances were also studied by Pervaporation Separation Index (PSI). These results are displayed in Table 2. From these results it is noticed that PSI values follow a decreasing trend with increasing amount of water in the feed. The % of water in permeate decreases with increase of mass % of water in feed. It is also observed that the % of water in permeate also increases with increase in zeolite concentration in the membrane. The pristine membrane % of water in the permeate is less than the zeolite-filled membranes.

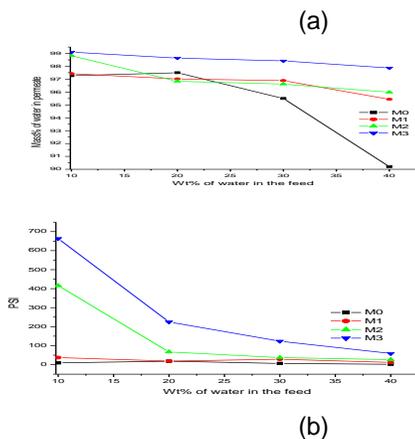


Figure : 8. a. Mass% of water in permeate Vs Mass % of water in feed for various percentages of MFI- 24Q zeolite filled NaAlg/HPC membrane for water-isopropanol mixtures at 30°C. **b.** Mass% of water in permeate Vs Mass % of PSI percentages of MFI- 24Q zeolite filled NaAlg/HPC membrane for water-isopropanol mixtures at 30°C.

3.1.9. Effect of feed composition on Pervaporation performance:

In pervaporation process, the overall selectivity of a membrane is generally explained based on the interaction among the membrane and permeating molecules, molecular size of the permeating species. **Figure: 9. (a)** displays the effect of water compositions on the flux for all the membranes prepared with different amounts of MFI-24Q zeolite in NaAlg/HPC matrix. It is observed that the flux of water in the feed increased drastically for all water/IPA

compositions under the studied zeolite concentration range (5-15 wt %). At higher concentration of water in the feed, the membranes swell greatly due to the formation of a strong interaction between the membrane and water molecules. This might have led to allow water molecules to pass through the membranes. [39] As a result, flux of water increases drastically at higher concentration of water in the feed, irrespective of the amount of zeolite loading in the membrane matrix. On the contrary, the selectivity decreased significantly from 10 to 40 upon increasing the water content in the feed. This can be clearly observed from **Figure : 9. (b)** showing the variation of selectivity as a function of wt% of water in the feed in the membrane for different mass % of MFI-24Q zeolite (5-15 wt %) in the membrane. In general, with increasing the water content in the feed compositions, the permeation flux increases and selectivity decreases. [28, 40] Similar observations were also observed in the present case with increasing water content in the feed composition. The highest flux observed for M3 is 0.864 kg/m²/h at a feed composition of 60/40 IPA/water mixture, where the highest selectivity observed is 991 at 10wt% of water in the feed.

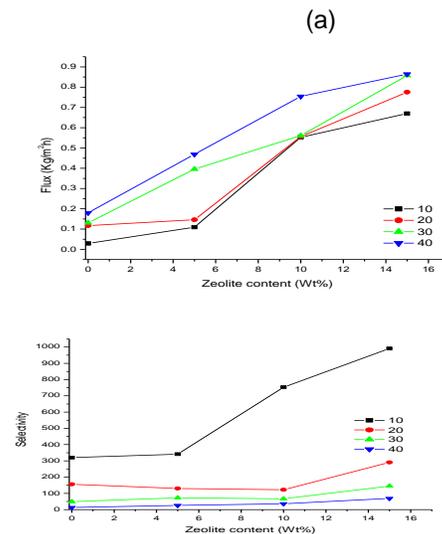


Fig : 9. (a) Effect of zeolite content on flux at different wt% of water in the feed. [■ -10, ● -20, ▲ -30, ▼ -40.] **(b)** Effect of zeolite content on selectivity at different wt% of water in the feed. [■ -10, ● -20, ▲ -30, ▼ -40.]

5. Conclusions:

MFI-24 Q Zeolite-filled NaAlg-HPC membranes were prepared for pervaporation separation of IPA-Water mixture, an increase of Zeolite content in the membrane results in a simultaneous increase of both flux and selectivity. This was explained on the basis of a significant increase in hydrophilic character, selective adsorption and molecular sieving action including reduction of pore size of the polymer matrix as evidenced by TGA, X-ray diffraction and Swelling studies. The Pervaporation separation index data also indicate that higher the degree of zeolite loading better is the membrane performance, indicating that the present MFI-24 Q zeolite loaded membranes are highly water selective. The present zeolite-filled membranes show the best and most stable PV separation characteristics for the

IPA-Water mixture, which in turn indicates that the membranes are highly hydrophilic and highly selective towards water. #This work is completed by the collaboration of the Dept of Chemistry and Dept of Polymer science & Tech., Sri Krishnadevaraya University, Anantapuramu, A.P.,INDIA.

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