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Article

Zeolite-enriched Blend Membranes of Sodium Alginate and Gelatin for Dehydration of Isopropanol by Pervaporation

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ABSTRACT

Blend membranes of anionic sodium alginate and cationic gelatin incorporated with H-mordenite zeolite were prepared by solution casting and solvent evaporation method for the dehydration of isopropanol solvent. The blend membrane containing 30% wt. of zeolite exhibited the highest separation selectivity of 1240 at a substantial flux of 0.64 kg/m^2 h for azeotropic feed composition at ambient temperature (30°C). The mixed matrix membranes were characterized by Fourier transform infrared spectroscopy, X-ray diffraction, thermal gravimetric analysis, and scanning electron microscopy to assess their intermolecular interactions, crystalline nature, thermal stability, and surface morphology, respectively. Extent of zeolite loading and effect of feed composition on the pervaporation performance of the membranes were evaluated. Experimental results showed that both flux and selectivity increased simultaneously with increasing zeolite content in the membrane. These observations were explained on the basis of enhancement of hydrophilicity, selective adsorption and molecular sieving action by the creation of selective openings in the blend matrix through zeolite incorporation.

Key words: Pervaporation, Sodium alginate-gelatin blend membrane, Isopropanol-water azeotrope, H-mordenite zeolite, Mixed matrix materials.

1. INTRODUCTION

Pervaporation (PV) allows isolation of valuable compounds from mixtures that are difficult to separate by distillation, extraction, and adsorption. PV 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 more permeable 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 performance of the organic polymer membranes. Hydrophilic zeolite membranes are capable of selective removal of water from its mixtures with organic compounds through PV. Zeolite membranes have uniform, molecular-sized pores and separate the feed molecules based on differences in their adsorption and diffusion properties. Zeolite membranes are thus well suited for separating liquid-phase mixtures by PV, and the first commercial application of zeolite membranes has been for dehydration of organics. Due to a 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 than 12 zeolite structures have been used in membranes [2,3]. H-mordenite zeolites are inorganic crystalline structures with uniform, molecular-sized pores and high mechanical, thermal and chemical stability making them suitable membrane-based applications [4-8]. Zeolite incorporation often increases both PV flux and selectivity [9] or increases separation factor with little or no decrease in flux [10-12]. However, polymeric systems that have exhibited improved separation factor and flux on inorganic filler incorporation include alcohol dehydration with poly (amide sulfonamide) and poly (vinyl alcohol) membranes filled with zeolite and halloysite [13,14].

Sodium alginate (SA), 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 exhibit excellent performance as a membrane material for PV -based dehydration of organic solvents [15,16]. Gelatin (GL) is a biodegradable, environmentally friendly “green” polymer. GL, which has been intensively studied in biomedical areas [17-19], is thought to be a green polymer candidate for membrane materials. It is well known that blending is an effective and convenient method to improve the performance of polymer materials. In the present study, blending of SA with GL has been explored for PV based separation of water from isopropanol (IPA). SA-GL blend solution was incorporated with H-mordenite in different ratios. The structure, morphology, thermal properties, and mechanical properties of these membranes were systematically investigated. Effects of H-mordenite zeolite content on

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the sorption and diffusion properties of the mixed matrix membranes were discussed. IPA is one of the important solvents used on a large scale in the chemical industry in 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 [19-21].

IPA is also more effective than ethanol for medical applications. IPA and water form an azeotropic mixture at 87.5 mass% of alcohol [22] and hence, the separation of this mixture by conventional methods such as solvent extraction, evaporation, or distillation could prove uneconomical for obtaining absolute IPA. Several membrane materials have been modified recently for the selective separation of water from aqueous mixtures of IPA [23]. 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 and separation factor in PV process. This study aims to improve the separation factor by the molecular sieving effect without any reduction in flux through use of mixed matrix membranes based on zeolite loaded SA-GL blends.

2. EXPERIMENTAL

2.1. Materials and Methods

2.1.1. Materials

SA was purchased from Loba Chemicals, Mumbai, India. GL, IPA, glutaraldehyde (GA), and hydrochloric acid were all purchased from s.d. Fine Chemicals, Mumbai, India. H-mordenite zeolite was supplied by Zeolite and Allied Products Pvt. Ltd., Mumbai. Deionized water generated by a reverse osmosis system in the laboratory was used throughout the research work.

2.1.2. Membrane preparation

SA (2 g) and GL (2 g) were individually dissolved in 100 ml of deionized water in two separate conical flasks with constant stirring for about 24 h at room temperature. The solutions were then filtered and mixed thoroughly in the ratio of 4:1 and cast onto a clean glass plate with the aid of doctor's blade and allowed to dry at room temperature for 1–2 days. The completely dried membrane was subsequently peeled-off and designated as SA-GL-0. To prepare H-mordenite filled SA-GL blend membrane, a known amount of H-mordenite zeolite was added into the SA/GL solution. The amount of SA-GL solution was kept constant each time. The zeolite filled blend solution was stirred for about 2 h, and then it was kept in an ultrasonic bath for 30 min at 30°C to break the aggregated crystals of H-mordenite zeolite and enhance its dispersion in the polymer matrix. The resulting solution was poured on to a glass plate, and the membrane was dried as mentioned above. The prepared membranes were then crosslinked in a bath containing with 84 vol. % IPA, 10 vol. % water, 5 vol. % of GA crosslinker, and 1 vol.% hydrochloric acid catalyst for a period of 2 h. The amount of H-mordenite zeolite with respect to SA-GL was varied 0, 10, 20, and 30 mass%, and the membranes thus obtained were designated as SA-GL-0, SA-GL-10, SA-GL-20, and SA-GL-30, respectively. The SA-GL blend was cross-linked with glutaraldehyde to reduce the extent of swelling. A model scheme of the possible interaction is shown in Figure 1 which represents the structure of SA-GL cross-linked with glutaraldehyde.

2.2. Membrane Characterization

All the prepared membranes used in PV measurements were characterized using the methods described below.

2.2.1. Fourier transform-infrared (FTIR) spectroscopic studies

The FTIR spectrum of SA-GL-0 and SA-GL-30 zeolite composite cross-linked membranes were scanned in the range between 4000 cm^{-1} and 400 cm^{-1} using Nicolet-740, Perkin-Elmer-283B FTIR (USA) Spectrometer by KBr pellet method.

2.2.2. X-ray diffraction (XRD) analysis

A Siemens D 5000 (Germany) X-ray diffractometer was used to study the solid-state morphology of SA-GL blend membranes and those incorporated with zeolites in different compositions. X-rays of 1.5406 A° wavelengths were generated by a CuK source. The angles of diffractions were varied from 2° to 65° to identify any changes in the crystal structure and intermolecular distances between intersegmental chains after modification.

2.2.3. Scanning electron microscopy (SEM) studies

SEM of surface and cross-section was taken for the SA-GL blend membrane and SA-GL zeolite filled membrane, using software controlled digital scanning electron microscope–JEOL JSM 5410, Japan.

2.2.4. Thermogravimetric analysis (TGA) studies

Thermal stability of the polymer films was examined (using Seiko 220TG/DTA analyzer, Japan) in the temperature range of 25–600°C at a heating rate of 10°C/min, with nitrogen flushed at 200 ml/min. The samples were subjected to TGA both before and after incorporation of zeolite to determine the thermal stability and decomposition characteristics.

2.2.5. Mechanical properties studies

Mechanical tests were performed at room temperature using the tensile testing machine (Shimadzu make and AGS-10 kNG model) with an operating head load of 5 kN. The cross-sectional area of the sample of known width and thickness was calculated. The films were then placed between the grips of the testing machine for a constant grip length of 5 cm. The speed of testing was set at the rate of 12.5 mm/min. Tensile strength (in N/mm^2) was calculated using the Equation 1.

$$\text{Tensile sterngth} = \frac{\text{Max load}}{\text{Cross sec tional area}} \tag{1}$$

2.2.6. PV experiments

The equipment used to perform the PV experiments remained the same as described earlier Sudhakar et al. [24,25]. 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 0.75 hp 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).

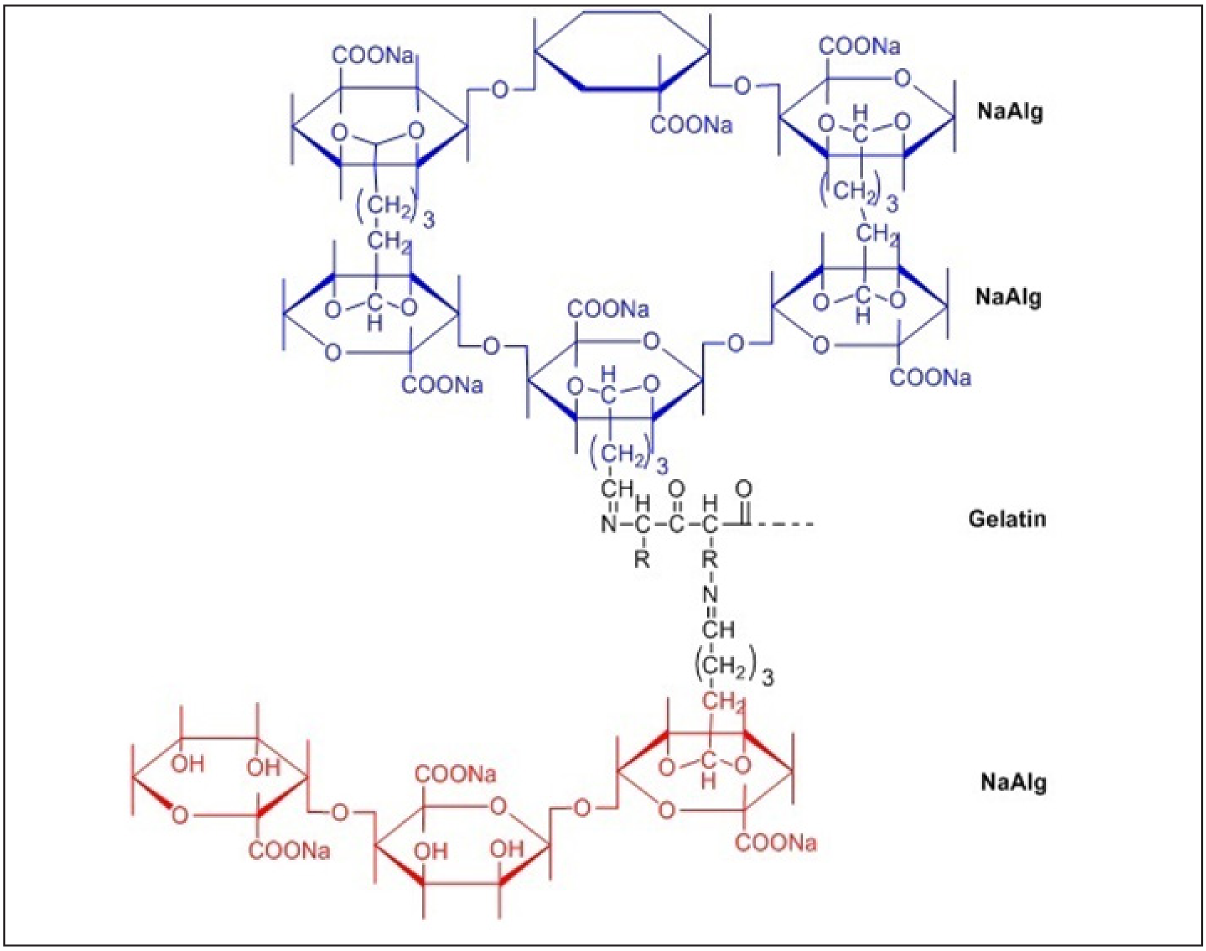

Figure 1: Reaction scheme of sodium alginate-gelatin blend cross-linked with glutaraldehyde.

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 a period of 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 (J_i) 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.

$$J_i = \frac{W_i}{A_t} \qquad (2)$$

Here W_i represents the mass of water in permeate (kg), A is the membrane area (m^2), and t represents the permeation time (h).

The composition of both feed and permeate composition was determined using a Nucon gas chromatograph (Model 5765) installed with thermal conductivity detector and packed column of 10% diethylene glycol sebacate on 80/100 Supelcoport of $1/8^{11}$ internal diameter and 2 m length. The oven temperature was maintained at 70°C (isothermal) while the injector and detector temperatures were maintained at 150°C each. The sample injection size was 1 µl, and pure hydrogen was used as the carrier gas at a pressure of 1 kg/cm^2. The GC response was calibrated for this particular column and conditions with known compositions of IPA-water mixtures and the calibration factors were feed into the software to obtain correct analysis for unknown samples, and the errors in the PV measurements were <1.0%. The selectivity of the H-mordenite zeolite-filled membranes was evaluated by Equation 3:

Membrane selectivity α, is the ratio of permeability coefficient of water to that of IPA, which is calculated from their respective wt. concentrations in feed and permeate as given below:

$$\alpha = \frac{y(1-x)}{x(1-y)} \qquad (3)$$

Where y is the permeate weight fraction of water and x is its feed weight fraction.

2.2.7. Sorption experiments

Interaction of the membranes with the pure liquid components of the feed mixture was determined by gravimetric sorption experiments. Dried membranes of known weight were immersed in different IPA-water mixtures for 48 h. When the sample attained constant weight, it was carefully wiped off with filter papers to remove surface liquid, and then quickly weighed. Degree of swelling was calculated as:

$$\text{Degree of swelling} = \frac{M_s}{M_d} \qquad (4)$$

Where M_s is mass of the swollen membrane in (g), and M_d is mass of the dry membrane in (g). The percent sorption was calculated using the equation:

$$\%\text{Sorption} = \left[\frac{M_s - M_d}{M_d}\right] \quad (5)$$

3. RESULTS AND DISCUSSION

3.1. Membrane Characterization

3.1.1. FTIR analysis

Figure 2 shows the FTIR spectra of the SA-GL-0 blend membrane (a) and SA-GL-30 composite cross-linked membranes (b). Both spectra show the carboxylic group at 1648 cm^{-1} characteristic peaks appearing in the range 3483–3413 cm^{-1} which correspond O–H stretching vibrations. This can be confirmed by the presence of a peak at 1095 cm^{-1} which corresponds to silanol.

The formation of a new peak (**Figure** 2a) at 1261 cm^{-1} corresponds to the interaction of hydroxyl group with GA crosslinker. The strong bond at 1726 cm^{-1} could be indicative of the free carboxyl group of SA. The bonds at 1618 cm^{-1} and 1413 cm^{-1} were assigned to the presence of the salified carboxyl group. In detail, 1618 and 1413 cm^{-1} bonds are assigned to the antisymmetric and symmetric COO^{-} stretching vibration of the salified carboxyl group; respectively, no direct evidence is available at present to assign the absorbance peak at 1231 cm^{-1} of SA membrane cross-linked with GA.

3.1.2. XRD

The WAXD patterns of pure SA/GL blend membrane and H-mordenite incorporated SA/GL blend membranes are presented in Figure 3. The XRD patterns of the membranes (b to d) show that as the extent of zeolite loading increased, peak intensity of the zeolite filled cross-linked membranes decreased compared to SA-GL-0 at around 2θ=20°. This revealed that the relative crystallinity of SA-GL cross-linked blend membranes increased with increasing loading of the zeolite. An additional peak also appeared as the zeolite content was increased at around 2θ=23°. This is due to the presence of H-mordenite zeolite in the membranes, whose intensity further increased as the zeolite content in the membranes increased from b to d.

3.1.3. SEM analysis

Figure 4a and b represent surface and cross-sectional SEM images of SA-GL membrane and SA-GL incorporated with H-mordenite zeolite, respectively. While the pure SA-GL-0 blend membrane shows a void-free dense structure, H-mordenite particles can be clearly observed in the SA-GL-10 to SA-GL-30 membranes. Much better homogeneity was obtained after zeolite incorporation as shown in Figure 4A (a-d), confirming an improvement incompatibility between the polymer and zeolite particles.

From the cross-sectional images displayed in Figure 4B (e-h), it could be mentioned that the SA-GL zeolite filled blend membrane displays better interfacial morphology than SA-GL-0 membrane. The two polymers and incorporated zeolite were homogeneously intermixed and in the state of a polymer composite membrane. No agglomerations are seen in the polymer blend before and after zeolite incorporation.

3.1.4. TGA studies

Figure 5 displays thermal degradation of SA-GL blend (SA-GL-0) and H-mordenite zeolite incorporated (SA-GL-10 to SA-GL-30) blend membranes by TGA. Two weight loss stages were observed in the temperature range between 140 and 550°C followed by the final decomposition of the SA-GL-0 membrane that began around 450°C.

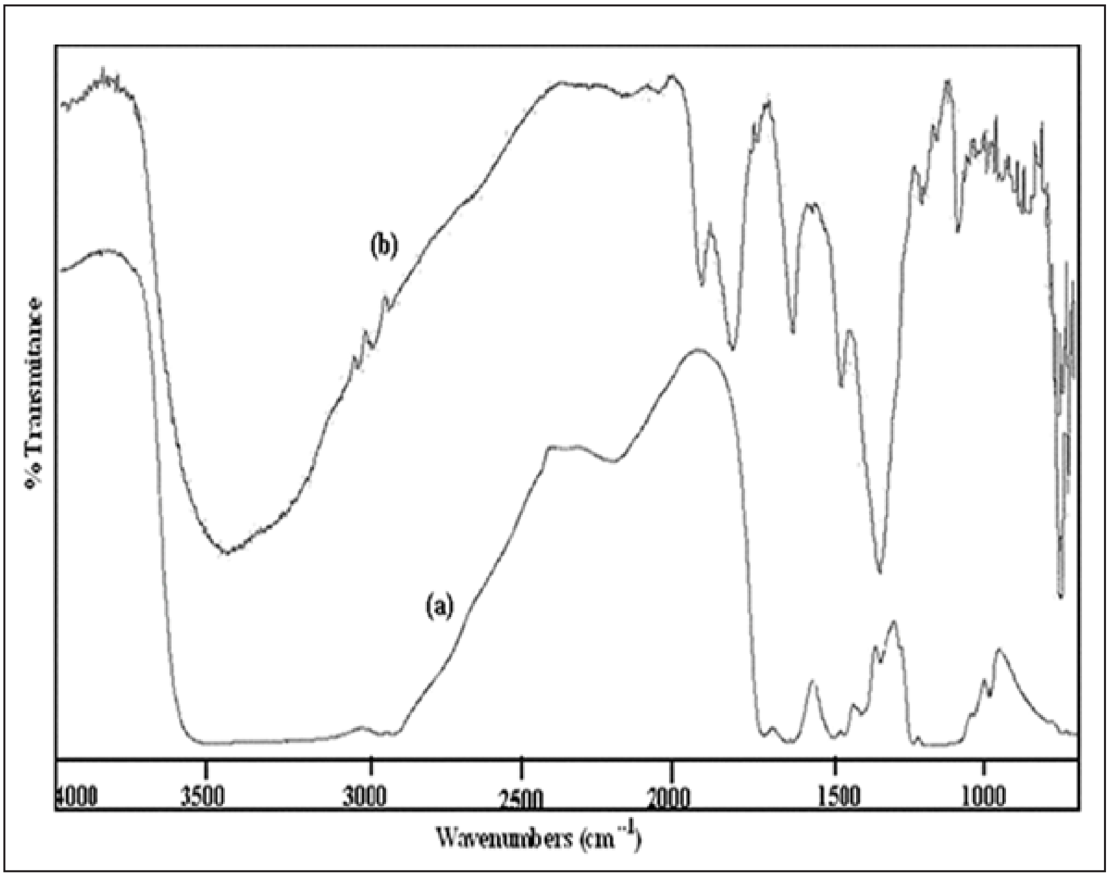

Figure 2: Fourier transform-infrared patterns of (a) sodium alginate-gelatin (SA-GL)-0; (b) SA-GL-30 cross-linked membranes.

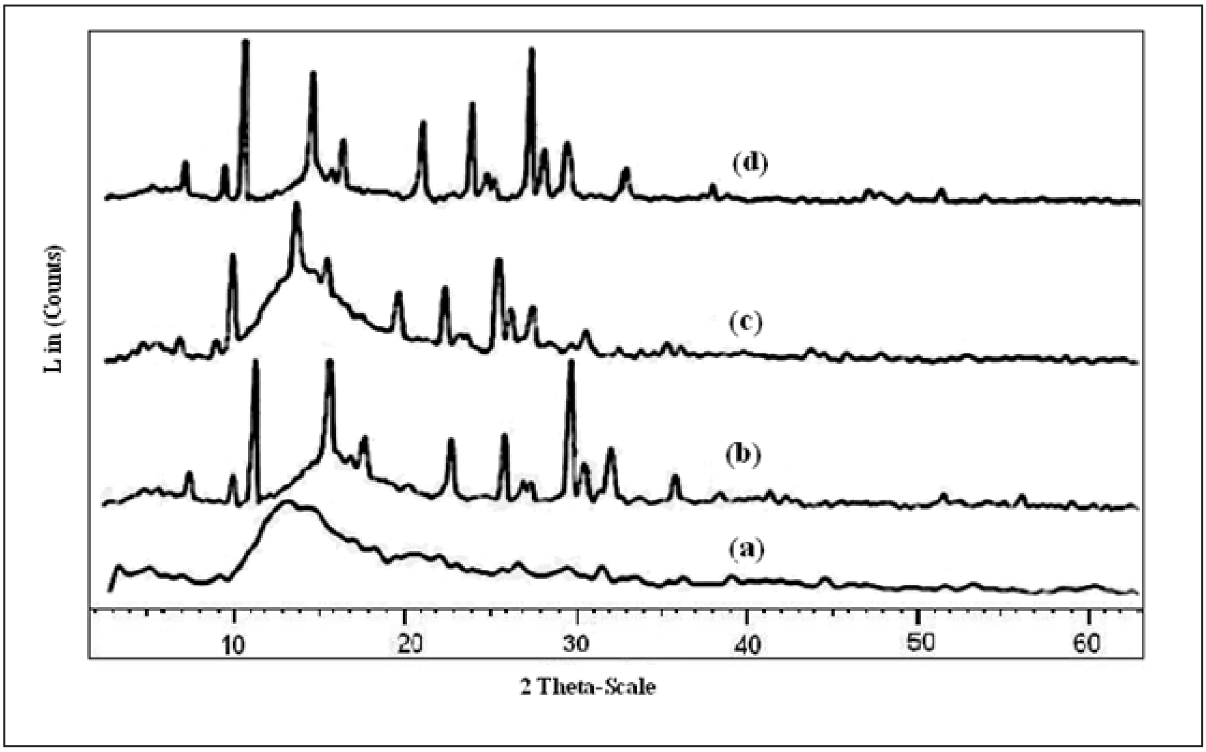

Figure 3: Wide angle X-ray diffraction patterns of zeolite filled sodium alginate-gelatin (SA-GL) blend membranes: (a) SA-GL-0; (b) SA-GL-10; (c) SA-GL-20; (d) SA-GL-30.

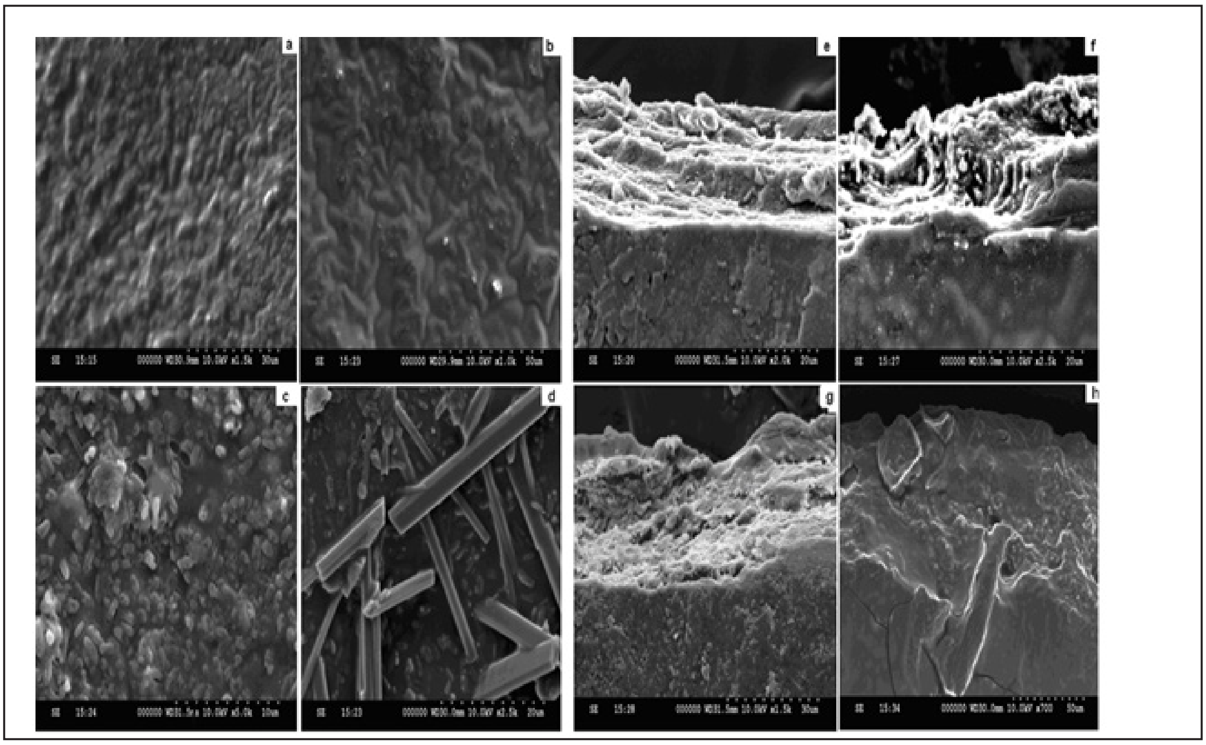

Figure 4: (A). Scanning electron microscopy (SEM) pictures surfaces of the: (a) Sodium alginate-gelatin (SA-GL)-0; (b) SA-GL-10; (c) SA-GL-20; (d) SA-GL-30. (B) SEM pictures of the (cross section): (e) SA-GL-0; (f) SA-GL-10; (g) SA-GL-20; (h) SA-GL-30.

Weight loss in the first stage could be attributed to splitting of the main polymer chains before its final decomposition.

In case of pure SA-GL blend membrane**,** two weight loss stages were observed around 140–230°C and 420–560°C followed by the final decomposition. After incorporating H-mordenite; the membranes (SA-GL-10 to SA-GL-30) exhibited two weight loss stages in the range of 180–260°C and 450–590°C followed by a final decomposition. An increase in thermal stability of the SA-GL H-mordenite zeolite blend was thus observed.

3.1.5. Mechanical properties

SA-GL-0, SA-GL-10, SA-GL-20**,** and SA-GL-30 cross-linked blend membranes exhibited tensile strengths of 9.14, 12.40, 14.80**,** and 15.74 N/mm^2, respectively, calculated from the stress–strain curves, which showed an enhancement in mechanical strength after incorporation of H-mordenite zeolite. The XRD spectra confirmed increased in crystallinity of SA-GL blend after incorporating H-mordenite, extensive intra**-** and inter-molecular hydrogen bonds of SA and GL, besides the formation of an interpenetrating polymer network on crosslinking, are responsible for such an increase in mechanical strength of the zeolite incorporated blends.

3.1.6. Membrane sorption properties

Preferential sorption is an important factor that decides the extent of separation. Different IPA-water compositions were used for sorption as well as separation studies. According to Figure 6, degree of swelling of the membrane increased as the water content in the feed solution increased. This is due to the fact that all the membranes are highly hydrophilic. Hence, water is preferentially absorbed and has a higher driving force than IPA to penetrate into the downstream side. It seems that the membrane with the highest content of H-mordenite exhibits maximum sorption due to sorption of water in the cylindrical pore structure of the zeolite particles.

3.1.7. PV properties

Hydrophilic zeolites are known to have a high affinity for water. As a result, there has been considerable work focused on trying to incorporate such zeolites as the active component in a membrane to obtain high flux and selectivity. Membrane performance was studied for dehydration of IPA at 30°C and displayed in Table 1 and Figure 7.

PV performance of the cross-linked SA-GL blend membrane and H-mordenite filled blend membrane was investigated by varying feed composition from 2 to 31 wt.% of water (Table 1). Increase in feed water content resulted in enhancement of flux but a reduction in selectivity for all the membranes due to the increased swelling in case of all the four membranes. The swollen membranes are plasticized and allow more and more transport of IPA molecules along with water molecules. From Figure 7, it can be noticed that when zeolite content is 30 wt**.**%, the flux generally is higher than that of the SA-GL-0 membrane, and the selectivity of the SA-GL-30 is higher especially at higher feed water concentrations. The effect of feed water concentration on the PV performance of the SA-GL-10 and SA-GL-20 membranes **is** similar to that of the SA-GL-30 membranes as shown in Figure 7.

3.1.8. Effect of H-mordenite zeolite loading

Zeolite filled SA-GL membranes were tested for the separation of IPA-water mixtures by PV which showed that flux and selectivity improved due to the molecular sieving effect of the zeolite even as its concentration was increased from 0 to 30 wt%. With increasing loading of zeolite particles, both selectivity and flux values have increased dramatically as shown Figure 8. Zeolites adsorb large quantities of water molecules in their fine pores due to preferential affinity. The presence of zeolite has, therefore, helped in creating more number of free channels to adsorb and the smaller sited transport water molecules with the simultaneous rejection of the larger sized organic component. A higher loading of zeolite has increased the selectivity of IPA-water mixtures due to the good compatibility between the polymer chains and the zeolite particles. 30 wt.% zeolite loaded composite membrane showed high selectivity and flux for IPA-water azeotropic mixtures.

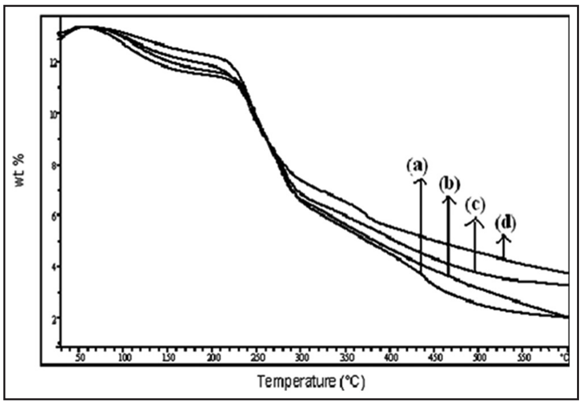

Figure 5: Thermogravimetric analysis of (a) sodium alginate-gelatin (SA-GL)-0; (b) SA-GL-10; (c) SA-GL-20; (d) SA-GL-30.

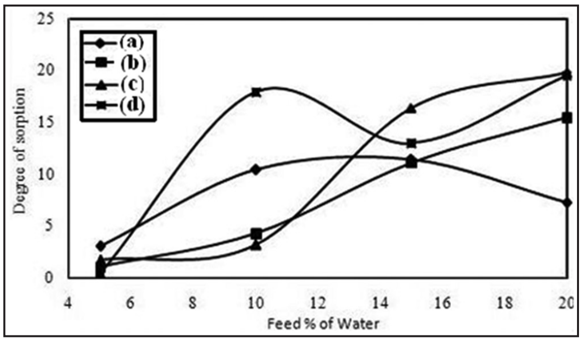

Figure 6: Plots of degree of sorption versus percentage of water in the feed (a) sodium alginate-gelatin (SA-GL)-0; (b) SA-GL-10; (c) SA-GL-20; (d) SA-GL-30 incorporated zeolite cross-linked membranes.

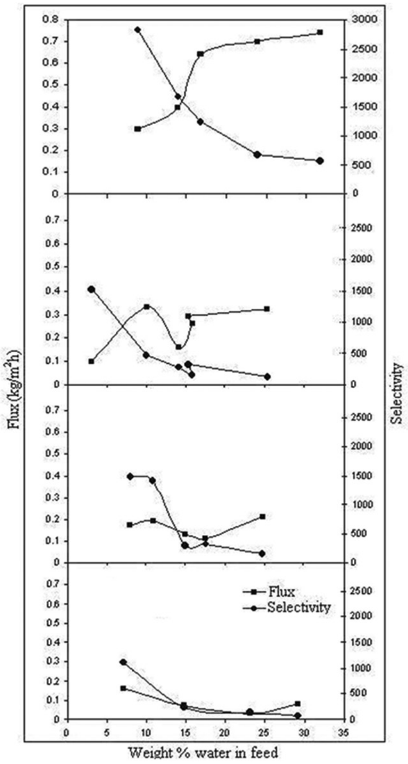

Figure 7: Effect of feed compositions on pervaporation performance of (a) sodium alginate-gelatin (SA-GL)-0; (b) SA-GL-10; (c) SA-GL-20; (d) SA-GL-30 incorporated zeolite cross-linked membranes.

Table 1: Pervaporation data of IPA and water mixtures at 30°C permeate pressure (1 mm Hg).

Feed compositions		Permeate compositions (wt.%)		Selectivity	Flux (kg/m^{2}h)
Water (x)	**IPA (1-x)**	**Water (y)**	**IPA (1-y)**		
SA-GL-0 membrane					
7.01	92.99	98.826	1.174	1116.66	0.16
14.771	85.229	97.644	2.356	241.18	0.07
23.013	76.987	96.374	3.626	131.06	0.03
29.226	70.774	86.235	13.765	75.62	0.08
SA-GL-30 membrane					
8.846	91.154	99.636	0.364	2820.65	0.30
13.923	86.077	99.632	0.368	1673.80	0.40
16.799	83.201	99.602	0.398	1239.45	0.64
23.923	76.077	97.632	2.368	672.97	0.70
31.799	68.201	92.602	7.398	569.22	0.74

SA-GL: Sodium alginate-gelatin, IPA: Isopropanol

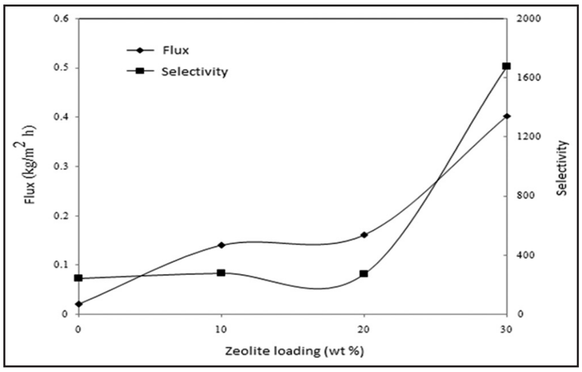

Figure 8: Zeolite concentration versus flux and selectivity of sodium alginate-gelatin blend membranes at azeotropic composition.

The present composite membranes are suitable for the achievement of >99% of water to the permeate side for IPA -water feed mixtures.

In general, all the mixed matrix membranes have shown better performance than the polymer blend. Thus, the unique properties such as molecular sieving and preferential adsorption come into effect only when the molecular sieve (zeolite) particles are compatible with the polymer phase (SA-GL). Hence, the interfacial properties of zeolite-filled polymer composites are critical for separation performance. The zeolite particles are able to discriminate between the components of a binary aqueous-organic mixture either by the exclusion of competing molecules on the basis of molecular size and shape or by adsorption preference. Zeolite has a pore size of 0.65 × 0.70 nm, which is larger than the kinetic diameter of water molecule (0.296 nm) and IPA (0.470 nm) [26] which was indeed the basis for selecting this zeolite to enhance the PV performance of SA-GL membrane since the larger IPA molecule will have restricted passage through the zeolite pore.

4. CONCLUSION

H-mordenite-filled SA-GL membranes were prepared for PV separation of IPA-water mixture. The developed membranes could break the azeotropic composition of IPA-water system. An increase of zeolite content in the membrane resulted 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. The PV separation data also indicate improved performance with increasing degree of zeolite loading. SEM studies showed uniform surface and cross-sectional morphologies which meant perfect blending of SA with GL as well as homogeneous distribution of zeolite fillers in the blends. X-RD showed **a** tighter structure with increased zeolite loading, and sorption studies revealed that the zeolite has preferential affinity for water which both substantiates the better selectivity observed. FTIR analysis confirmed the cross-linking of both SA and GL polymers by glutaraldehyde; whereas tensile testing and TGA confirmed that zeolite incorporation improved mechanical stability and did not affect thermal properties adversely. This H-mordenite zeolite loaded SA-GL blends appear to be promising candidates for separation of IPA-water azeotrope.

5. ACKNOWLEDGMENT

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