Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Ahmad, A. L. , Nawawi, M. G. Mohd. and So, L. K.(2005) 'Development of Novel NH_4Y Zeolite-Filled Chitosan Membranes for the Dehydration of Water-Isopropanol Mixture Using Pervaporation', Separation Science and Technology, 40: 15, 3071 — 3091

To link to this Article: DOI: 10.1080/01496390500385111

URL: http://dx.doi.org/10.1080/01496390500385111

Separation Science and Technology, 40: 3071–3091, 2005

ISSN 0149-6395 print/1520-5754 online
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Development of Novel NH_4Y Zeolite-Filled Chitosan Membranes for the Dehydration of Water-Isopropanol Mixture Using Pervaporation

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Abstract: NH_4Y zeolite-filled chitosan membranes were developed for the separation of water-isopropanol mixture using pervaporation process. The NH_4Y zeolite-filled chitosan membranes were prepared using a solution technique with the variation of NH_4Y zeolite loading (0, 0.1, 0.2, 0.3, 0.4, 0.5 wt.%). The membranes morphologies were studied using Scanning Electron Microscopy (SEM) and the membranes mechanical strength were tested using the parameter of tensile strength and percent strain at maximum. The effects of NH_4Y zeolite loading on the liquid sorption characteristics and pervaporation performance were also evaluated. The diffusion coefficient of water and isopropanol for the chitosan membranes at different NH_4Y zeolite loading is estimated. The presence of NH_4Y zeolite in the chitosan membranes caused non-homogeneous dispersion of NH_4Y zeolite crystals and membrane swelling due to its hydrophilic properties. However, the presence of NH_4Y zeolite was able to improve both tensile strength and percent strain at maximum of chitosan membranes. The presence of NH_4Y zeolite also increased the total permeation flux and separation factor simultaneously. The Pervaporation Separation Index shows that 0.2 wt.% of NH_4Y zeolite-filled membrane gave the optimum performance in the pervaporation

Received 15 February 2005, Accepted 25 August 2005

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process. The diffusion coefficient estimated proves that the membranes were highly water selective.

Keywords: Pervaporation, zeolite, chitosan, water-isopropanol

INTRODUCTION

Pervaporation is typically suited to separating a component of a liquid mixture to obtain high purity liquid. The pervaporation process has attracted increasing attention as an effective and energy-efficient hybrid separation process for the separation of azeotrope or close-boiling liquid mixtures (1). Isopropanol-water mixture forms an azeotrope in 14.7 mass% of water (1) and thus conventional distillation is not feasible. The pervaporation process is an alternative to providing a promising technique in the separation of isopropanol-water mixture because it can be used to separate the liquid mixture over the entire range of concentration irrespective of the presence of azeotrope.

Hydrophilic membranes are used for the dehydration process to remove water from organic solutions. The common hydrophilic membrane are poly(amide sulfonamide) and poly(vinyl alcohol) (2), sodium alginate (3), polyimide (4) and chitosan (5). Chitosan (poly[β-(1 $\rightarrow$ 4)-2-amino-2-deoxy-D-glucopyranose]) is a derivative of chitin or obtained through N-deacetylation process (6). Chitin is the second abundant natural biopolymer after cellulose. Sources of chitin are the protective shells of crustaceans such as crabs, shrimp, and lobster (7). The chitosan membrane is found to have favorable permselectivity and solvent stability, good film forming properties, highly hydrophilic and good chemical resistant properties (8).

Ge et al. (9) had used the chitosan membrane in the pervaporation process to separate the ethanol-water mixture and found that the chitosan membrane is the most effective water-permselective material due to the membrane high separation factor. It can be used for homogeneous or skin layer of composite membranes. However, the larger free volume between the molecular chains of pure chitosan membrane cause the separation performance not satisfactory in terms of total permeation flux, separation factor and mechanical strength (10).

Several researchers have modified the chitosan membrane for a better dehydration performance. Tang et al. (11) improved the membrane performance by blending the pure chitosan with the PVA polymer. Chen et al. (12) have reported the preparation and characterization of HY zeolite-filled chitosan membranes for the dehydration of organic-water mixtures using pervaporation. A flux of $353\,\mathrm{g/m^2 \cdot h}$ and a selectivity of 102 were obtained for an ethanol-water mixture with 10 wt.% of water at room temperature when the HY zeolite content in the membrane was 20 wt.%. This improved performance was due to the high hydrophilicity (higher alumina to silica ratio) and strong interaction between HY zeolite and chitosan.

Kariduraganavar et al. (3) had improved both permeation flux and separation factor simultaneously by modifying the sodium alginate membrane with the incorporation of NaY zeolite. The membrane containing 30 mass% of NaY zeolite was found to have the highest separation selectivity of 614.33 with a flux of $14.59 \times 10^{-2}\,\mathrm{kg/m^2\,h}$ for 5 mass% of water in the feed mixture at 30°C.

The incorporation of hydrophilic zeolite or porous filler into the dense membrane can improve the separation performance of the membrane. Synthetic zeolites have high crystalline ordered structure and hence offer the unique advantage of narrow pore size distribution. Thus the zeolite-filled membrane is able to show higher molecular sieving properties, selective adsorption and difference in diffusion rates. In addition, zeolites have a high mechanical strength, good thermal and chemical stability, and the membranes incorporated with these fillers can be used over a wide range of operating conditions due to their inorganic nature (3). Therefore, the NH_4Y zeolite is chosen as the chitosan membrane filler in the present studies due to its hydrophilic characteristic.

The objective of the present study is to enhance the permeation flux, separation factor, and mechanical strength simultaneously by modifying the water selective chitosan membranes with the incoparation of NH_4Y zeolite as the membranes filler using a solution technique. The NH_4Y zeolite-fill chitosan membranes performance at difference NH_4Y zeolite loading (0.1 wt.%–0.5 wt.%) is also evaluated for the dehydration of isopropanol-water mixture using pervaporation. The diffusion coefficient of water and isopropanol for the chitosan membrane with different NH_4Y zeolite loading is calculated by using the pervaporation data. The diffusion coefficient data can further improve understanding of the transport mechanism in pervaporation.

EXPERIMENTAL

Materials

Chitosan polymer (MW: 50,000–100,000) from UNZA Pharmaceutical, Malaysia was used. Reagent grade acetic acid was supplied from Mallinckrodt Baker. NH_4Y zeolite with average particle size ~2 μm in powder form was purchased from Aldrich Chemical Company, USA. Isopropanol was purchased from Merck, Germany. Distilled water was used throughout the research work.

Preparation of Chitosan Membrane

A pre-weighed chitosan powder was dissolved in 10 wt.% aqueous acetic acid solution for 24 hours at room temperature to produce a casting solution consisting of 2 wt.% chitosan. The polymer solution was filtered and the solution was left overnight at room temperature. The casting solution was cast onto a petri dish and allowed to spread slowly and evaporate in a dust-free atmosphere for 3–4 days at room temperature. The formed membranes were peeled off from the plate. 3 wt.% of NaOH solution containing 47 wt.% ethanol and 50 % deionized water was used to treat the formed membranes at room temperature for 24 hours. The formed membranes were washed thoroughly with deionised water and dried at room temperature.

The NH_4Y zeolite-filled chitosan membranes were prepared by adding a known amount of NH_4Y zeolite into the casting solution. The amount of the chitosan powder used was kept constant. The casting solution was cast, dried, and treated as mentioned above. The quantities of NH_4Y zeolite had been used were varied as 0.1, 0.2, 0.3, 0.4, and 0.5 wt.%.

Pervaporation Experiments

Pervaporation schematic diagram is shown in Fig. 1. The effective area for membrane permeation was 46.6 cm^2 and the glass feed tank had a feed

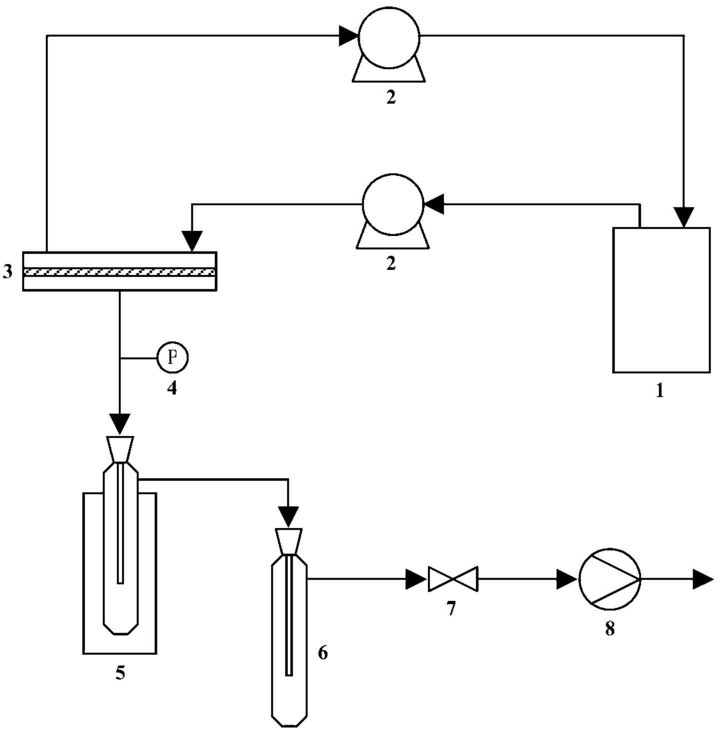

Figure 1. Schematic representation of pervaporation apparatus: (1) feed tank; (2) circulation pump; (3) membrane cell; (4) pressure gauge; (5) permeate cold trap; (6) moisture cold trap; (7) control valve; (8) vacuum pump.

solution capacity of approximately 1 L. The water composition in the feed was kept constant at 10 wt.%. The feed mixture was circulated through the cell by a Masterflex peristaltic circulation pump with the concentrate recycled back to the feed tank. The upstream pressure was maintained at atmospheric pressure. The vapor permeated through the membrane was withdrawn from the lower part of the cell, which was kept under vacuum with a vacuum pump and condensed in a cold trap that was immersed in liquid nitrogen. The control valve was used to control the downstream pressure at 60 mmHg. The pervaporation was run for at least 1 hr to get the system up to a steady state before condensing the collection. Permeate was warmed up to ambient temperature, weighed, removed, and analysed for permeate composition. The composition of permeate was analysed using a refraction index. The feed composition was assumed to be constant during the duration of the experiment due to the weight of permeate collected was less than 1% of the weight of the feed charged to the tank. In all experiments the temperature of the permeate cell was at room temperature.

The membranes performance in pervaporation was characterized by:

Total flux, J_t

$$J_t = \frac{W}{\Delta t A} \tag{1}$$

Separation factor, $\alpha_{w/IPA}$

$$\alpha_{w/IPA} = \frac{y_w/y_{IPA}}{x_w/x_{IPA}} \tag{2}$$

Enrichment factor, β

$$\beta = \frac{\mathrm{y_w}}{\mathrm{x_w}} \tag{3}$$

Pervaporation Separation Index, *PSI*

$$PSI = J_t(\alpha_{w/IPA} - 1) \tag{4}$$

where W is the weight (g) of permeate, Δt is the permeation time (h), A is the membrane area (m^2), x and y are the weight fraction of the feed and permeate respectively while the subscripts w and IPA denote water and isopropanol respectively.

Scanning Electron Microscopy (SEM)

The morphology of the membranes were determined by PHILIPS XL-40 scanning electron microscope at 15 kV accelerating voltage. The membranes were freeze-fractured in liquid nitrogen and then mounted on the aluminium stub. The specimens were sputter-coated with gold prior to macroscopic observation. The membranes were examined to determine if the zeolite particles were dispersed homogenously and if there were any defects or flaws in the membranes through the surface and cross section SEM micrographs.

Mechanical Strength of the Membranes

The tensile strength and the percent strain at maximum tests for the membranes were performed with LLYORD EZ 50 instrument. The membranes were cut into a dumbbell shape. The width and thickness of specimens at several points along their narrow sections were measured with an electronic digital calliper and digimatic micrometer. Then, the specimens were placed in the grips of the testing machine and the extension indicator was attached. The crosshead speed was fixed at 10 mm/min for tensile strength and percent strain at maximum test. The minimum specimens needed for each sample were five (STD. ASTM D638-ENGL, 1999).

Tensile Strength

$$\textit{Tensile Strength} = \frac{\textit{Load at Maximum}}{\textit{Area of Membrane Sample}} \tag{5}$$

Percent Strain at Maximum

$$\textit{Percent Strain at Maximum} = \frac{\textit{Extension at Maximum}}{\textit{Gauge Length}} \times 100 \tag{6}$$

Liquid Sorption Experiments

Degree of Swelling (DS)

The pre-weighed membranes after keeping the membranes in desiccators to desorb any moisture absorbed from the air were immersed in a known composition of isopropanol-water mixtures in a closed bottle at room temperature for 48 hours for an equilibrium swelling. The swollen membranes were removed and weighed immediately after being pressed between tissue papers. The amount of sorbed liquid in the membranes was expressed as the degree of swelling (*DS*), which was calculated using

$$DS\ (\%) = \frac{(w_f - w_i)}{w_i} \times 100\ (\%) \tag{7}$$

where w_f and w_i denote the final (swollen) and the initial (dry) weight of the membranes respectively.

Sorption Selectivity ($\alpha_{sorption\ w/IPA}$)

The membranes were then immediately placed in a desorption cell that was connected to a cold trap followed by a vacuum pump. The liquid sorbed by the membranes were desorbed under vacuum and collected in the trap. The collected liquid was then weighed, removed, and analysed for composition using the measurement of refraction index. The membranes were periodically weighed until the mass had been constant. The individual sorbed amount was calculated from the total sorbed amount and the sorbed composition. Sorption selectivity ($\alpha_{sorption\ w/IPA}$) was calculated as follows (13).

$$\alpha_{sorption\ w/IPA} = \frac{C_w/C_{IPA}}{X_w/X_{IPA}} \tag{8}$$

C_w and C_{IPA} are the weight fraction of the permeate water and isopropanol component in the membranes at the equilibrium sorption. $X_w$and X_{IPA} are the weight fraction of the feed water and isopropanol component at equilibrium sorption respectively.

RESULTS AND DISCUSSION

Effects of NH_4Y Zeolite Loading on Membrane Morphologies

The cross-sectional morphology and the bottom surface view of the membranes were shown in Figs. 2 and 3 respectively with the level of magnification of 2000× (except the bottom surface view of 0 % NH_4Y zeolite which is 4000×). Figures 2(a) and 3(a) show that the homogeneous chitosan membrane (0 wt.% of NH_4Y zeolite) had an uniform structure and no pores were visible even at the high level of magnification. This shows that the homogeneous chitosan membrane was a dense membrane (14).

Figure 2(b)–2(f) show that non-homogenous dispersion of the NH_4Y zeolite crystals in chitosan membranes was obtained. This resulted from the precipitation of NH_4Y zeolite at the bottom layer of the membranes. This was caused by the difference density of inorganic nature of NH_4Y zeolite in the chitosan membrane (15). Aggregation of NH_4Y zeolite can be observed at the bottom surface view of the membranes from Fig. 3(b)–3(f). The NH_4Y zeolite was not completely covered by the chitosan polymer phase. As a result, pinholes or non-selective voids were formed (16).

However, the aggregation of NH_4Y zeolite in chitosan polymer was still fine and the dispersion of the NH_4Y zeolite was the most homogeneous at 0.2 wt.% loading (Fig. 3(c)) compared to the other NH_4Y zeolite loadings. NH_4Y zeolite was dispersed well in the chitosan polymer and non selective voids were covered by the chitosan polymer. During the membrane

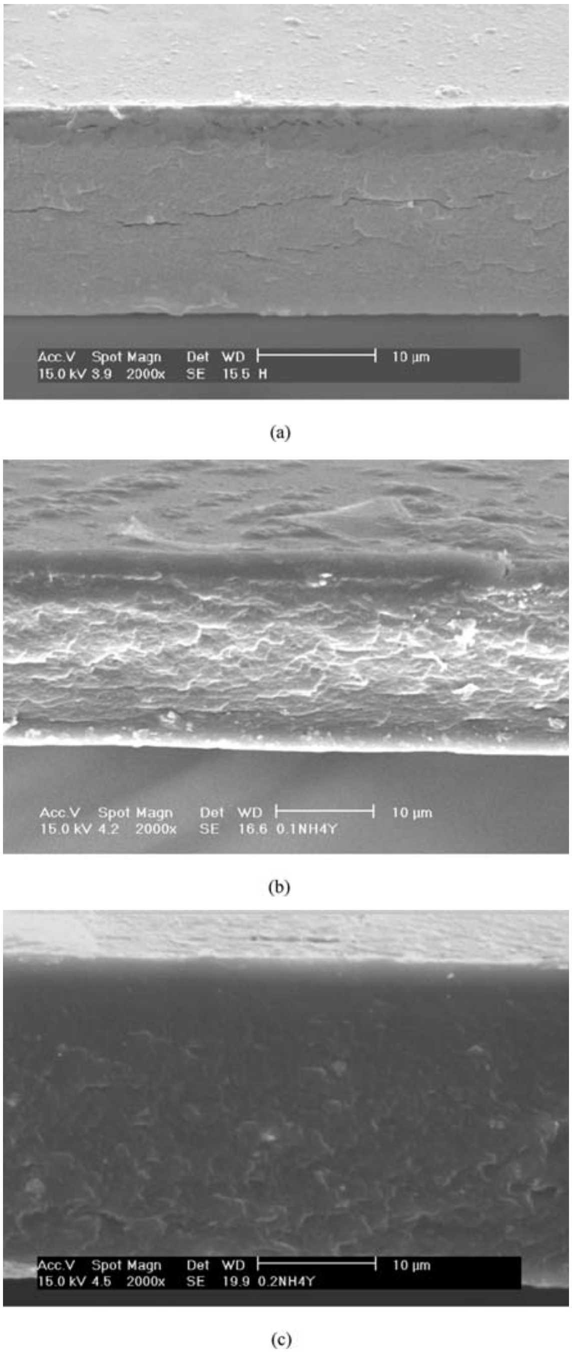

Figure 2. Cross-sectional morphology of (a) 0 wt.%, (b) 0.1wt. %, (c) 0.2 wt.%, (d) 0.3 wt.%, (e) 0.4 wt.%, (f) 0.5 wt.% zeolite NH_4Y-filled chitosan membrane.

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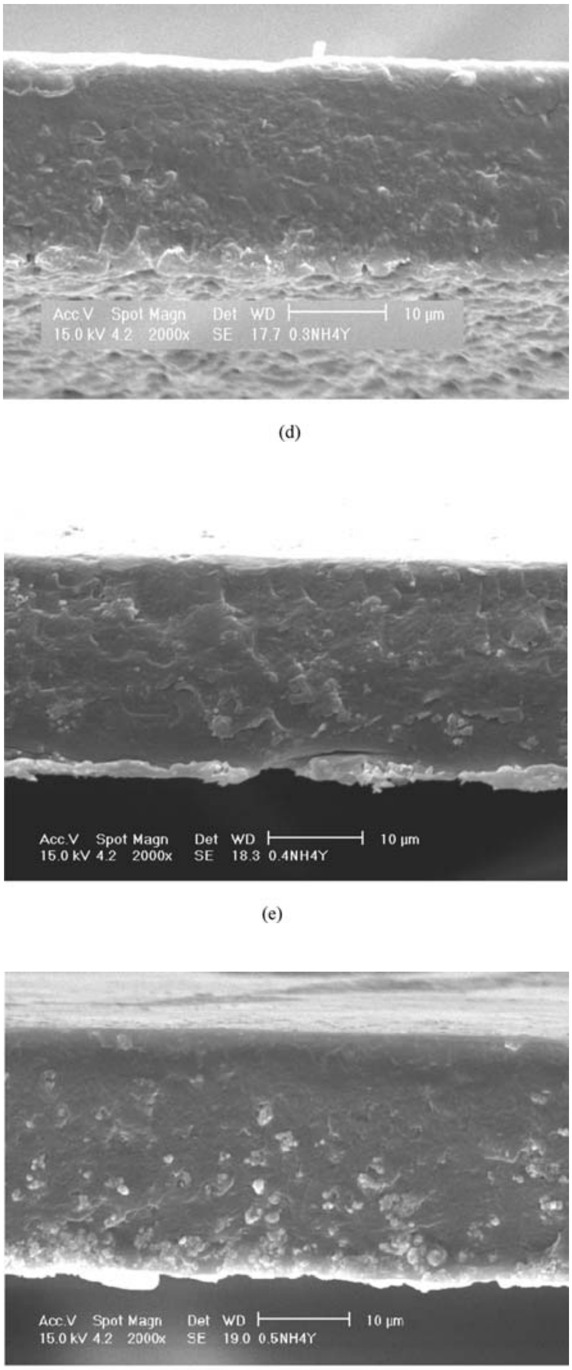
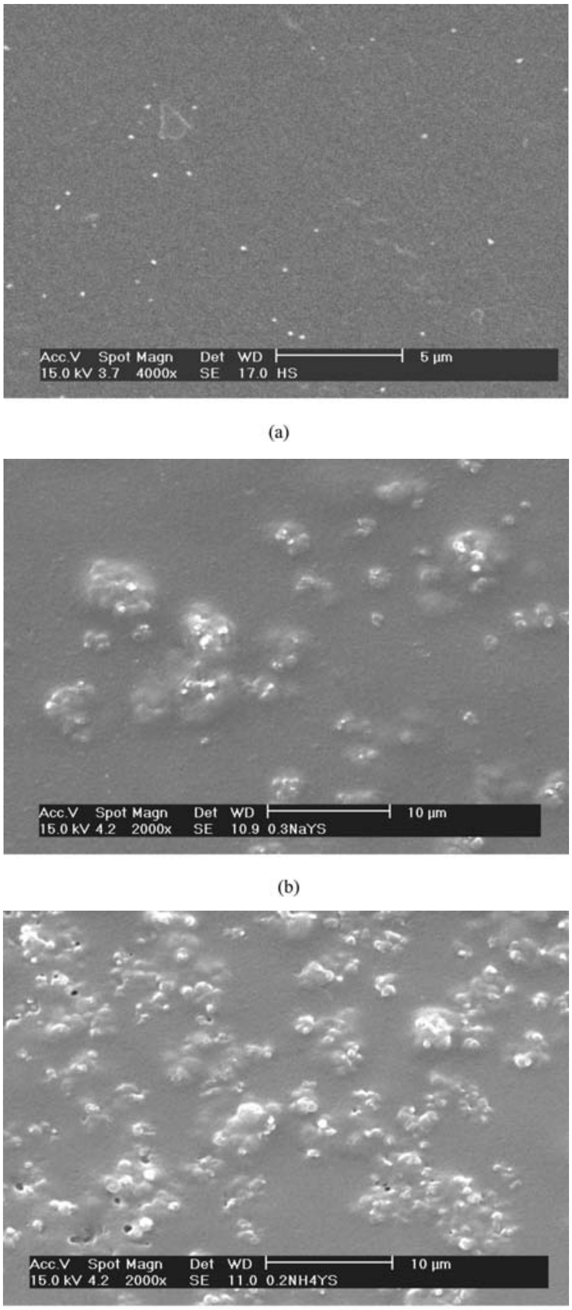


(d)

(e)

(f)

Figure 2. Continued.

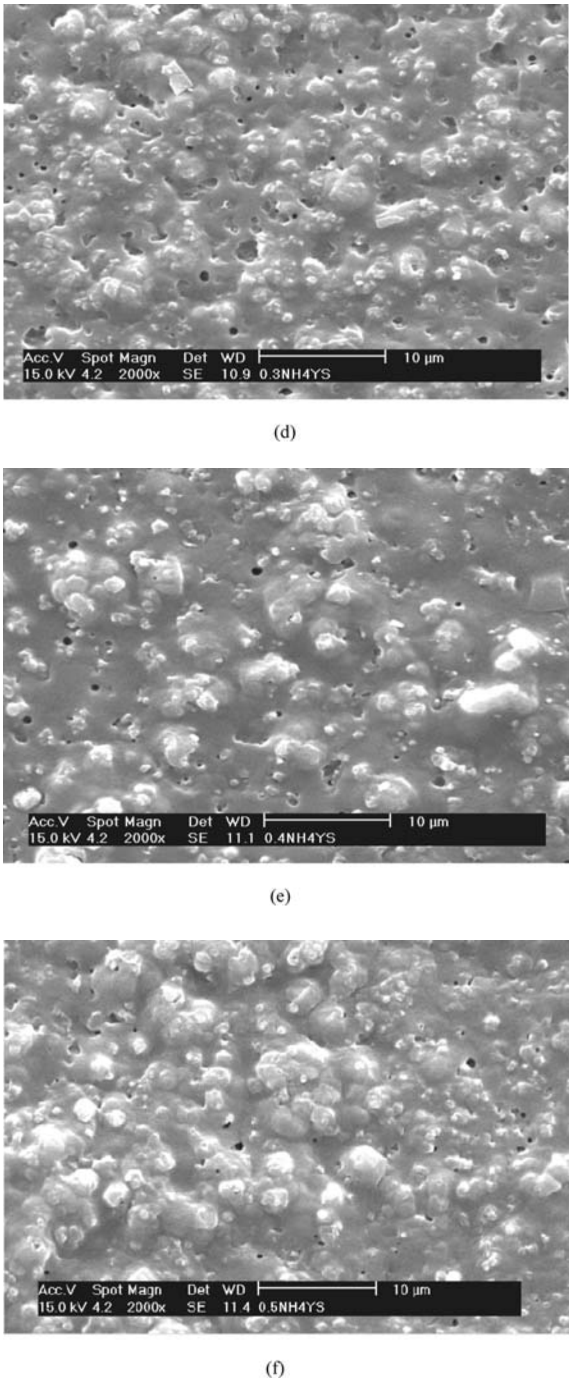


(a)

(b)

(c)

Figure 3. Bottom surface view of (a) 0 wt.%, (b) 0.1 wt.%, (c) 0.2 wt.%, (d) 0.3 wt.%, (e) 0.4 wt.%, (f) 0.5 % zeolite NH_4Y-filled chitosan membrane.



(d)

(e)

(f)

Figure 3. Continued.

preparation, 0.2 wt.% loading gave the least precipitation of NH_4Y zeolite compared to other membranes of different NH_4Y zeolite loading.

Effects of NH_4Y Zeolite Loading on Membrane Mechanical Properties

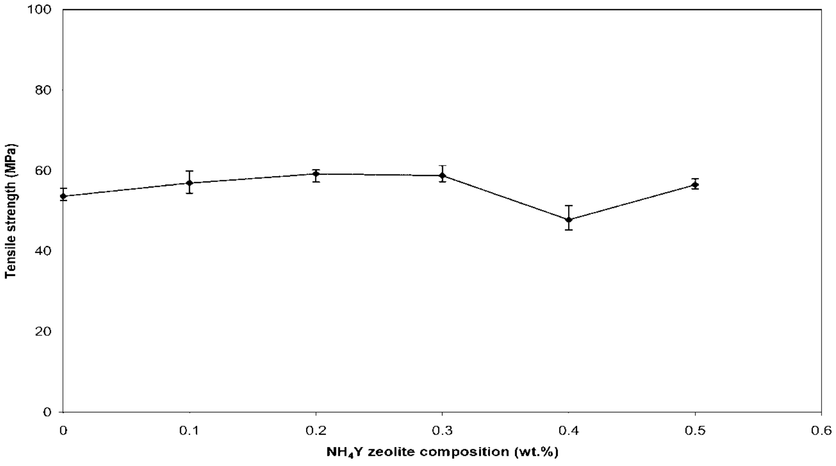
Figure 4 shows NH_4Y zeolite was able to improve both tensile strength and percentage of strain at maximum for chitosan membranes using the NH_4Y zeolite. From Fig. 4(a) the maximum tensile strength of the membrane was obtained at 0.2 wt.% of NH_4Y zeolite loading and the tensile strength start to decrease from 0.3 wt.% of NH_4Y zeolite loading. This shows that NH_4Y zeolite was not dispersed well at loading higher than 0.2 wt.% and the effect of aggregation resulted the interaction between NH_4Y zeolite and chitosan became weaker to extend for the load at maximum.

The strain at maximum increased when the NH_4Y zeolite loading increase is shown in Fig. 4(b). The highest percentage of strain at maximum was achieved at 0.4 wt.% of NH_4Y zeolite loading. The strain at maximum increased at increasing NH_4Y zeolite loading due to the chains mobility where the chains between NH_4Y zeolite and chitosan polymer were not well joined together caused by the aggregation of NH_4Y zeolite. Therefore, the ability of the membrane to extend for the load at maximum for tensile strength test will be decreased. Based on the observed results of Figs. 2, 3, and 4, the loading of NH_4Y zeolite for more than 0.5 wt.%, the tensile strength and percentage of strain at maximum will decrease.

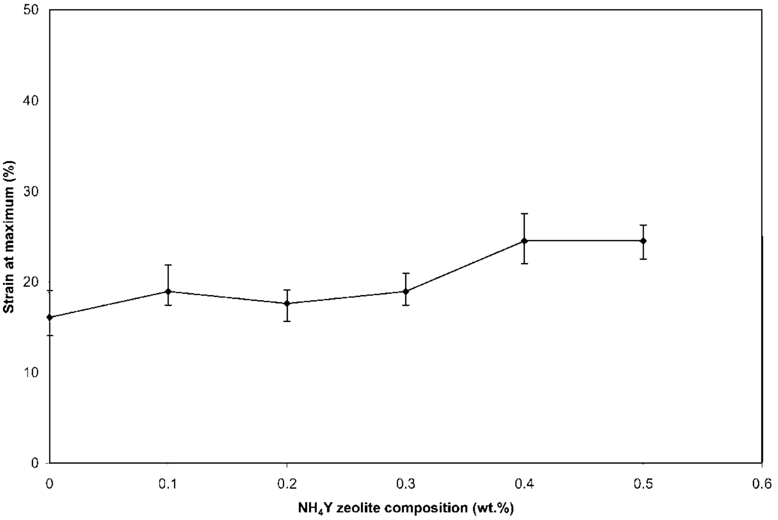
Effects of NH_4Y Zeolite Loading on Membrane Swelling and Sorption Selectivity

The degree of swelling and sorption selectivity for NH_4Y zeolite-filled chitosan membrane are shown in Fig. 5. It was observed that when the NH_4Y zeolite loading increased the degree of swelling is also increased. However, the degree of swelling decreased when the NH_4Y zeolite loading was 0.5 wt.%. The loading of NH_4Y zeolite increased the degree of swelling due to its hydrophilic properties (because the NH_4Y zeolite increase the membranes ability to absorb water) but the increments were very small because of the inorganic properties of NH_4Y zeolite which resisted to the swelling effect (15).

Figure 5 shows that the sorption selectivity decreased when the NH_4Y zeolite loading increased. However the water absorbed by the membrane was always higher than the isopropanol. This suggested that there were different degrees of polymer-permeate interaction between chitosan with water and isopropanol molecules. In addition, chitosan polymer is highly hydrophilic (8).



(a)



(b)

Figure 4. Mechanical properties of membrane at different NH_4Y zeolite loading. (a) Tensile strength; (b) strain at maximum.

Effects of NH_4Y Zeolite Loading on Pervaporation

Figure 6 shows that the total permeation flux, water, and isopropanol permeation flux increased to the maximum at 0.3 wt.% of NH_4Y zeolite loading. Modification of the chitosan membrane using the NH_4Y zeolite improved the total, water and isopropanol permeation flux because the introduction of

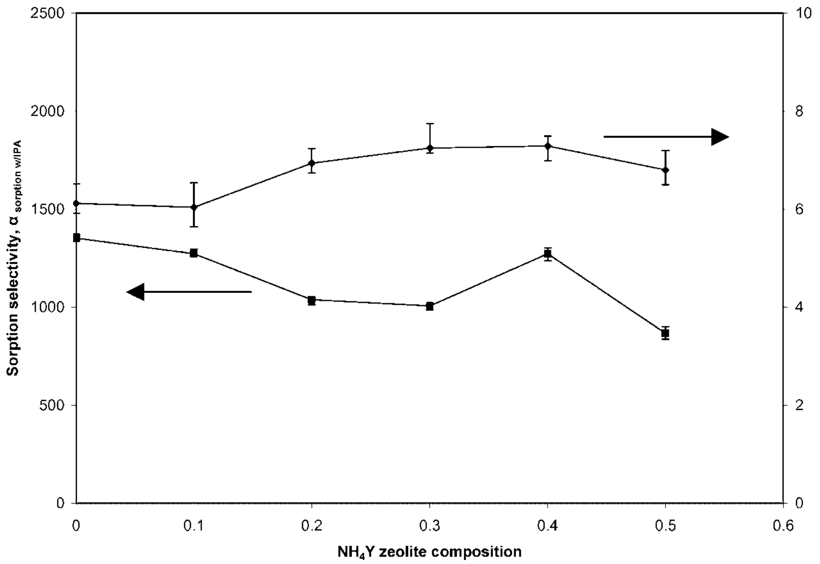


Figure 5. Variation of degree of swelling and sorption selectivity at different NH_4Y zeolite loading.

NH_4Y zeolite with smaller particle size and higher specific pore volume leads to a decreased in the diffusion resistance during pervaporation (15). Although the total, water and isopropanol permeation flux value is higher than the value of homogeneous chitosan membrane (0 wt.% of NH_4Y zeolite), the value of

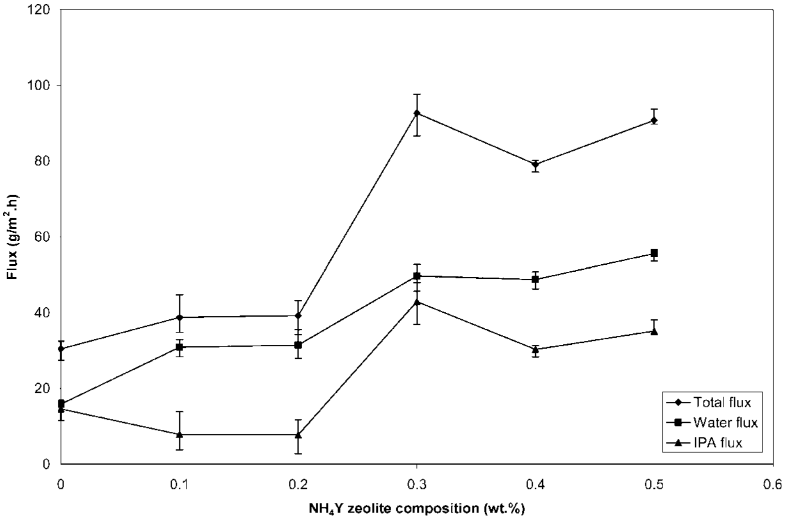


Figure 6. Effect of different NH_4Y zeolite loading on the total, water and IPA flux at 10 wt.% of water in the feed.

the fluxes started to decrease from 0.3 % of NH_4Y loading. At this region, the water molecules were difficult to pass through the tiny pores of NH_4Y zeolite and the permeation took place only in the polymer matrix of the membranes. Therefore, the dependence of the facilitation permeation on the molecular size could not longer be observed (17).

The separation factor increased to the maximum at 0.2 wt.% of NH_4Y zeolite loading as shown in Fig. 7. This was due to the high water selectivity of the NH_4Y zeolite and the stability of the membrane (high crystallinity of chitosan strongly resists the swelling effect). When the NH_4Y zeolite loading was more than 0.2 wt.%, the separation factor started to decreased. During the pervaporation process, when water and isopropanol molecules diffused through the chitosan membrane, the crystallinity of chitosan decreased at high NH_4Y zeolite loading and the membrane became swollen. The swelling process decreased the NH_4Y zeolite-chitosan interaction and resulted some tiny cracks at the NH_4Y zeolite-chitosan interface (6). Thus, free volume increased due to the plasticity effect and more isopropanol molecules were allowed to pass through the membrane (8).

Figure 8 shows that the NH_4Y zeolite loading of 0.2 wt.% achieved the highest Pervaporation Separation Index. The total flux and separation factor of 0.2 wt.% NH_4Y zeolite-filled chitosan membrane were 1.3 and 3.8 times higher than the homogeneous chitosan membrane respectively. The Pervaporation Separation Index was also 5.3 times higher than the homogeneous chitosan membrane. Therefore, the present studies show that 0.2 wt.% of

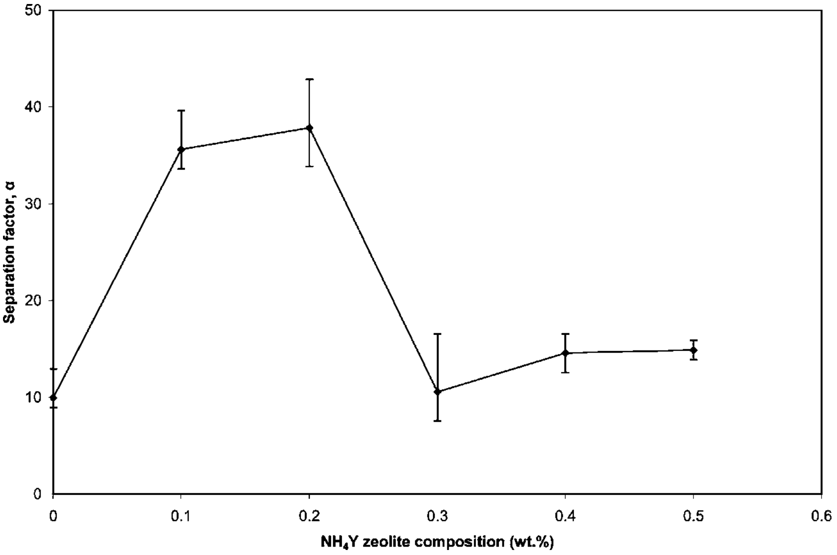


Figure 7. Effect of different NH_4Y zeolite loading on separation factor at 10 wt.% of water in the feed.

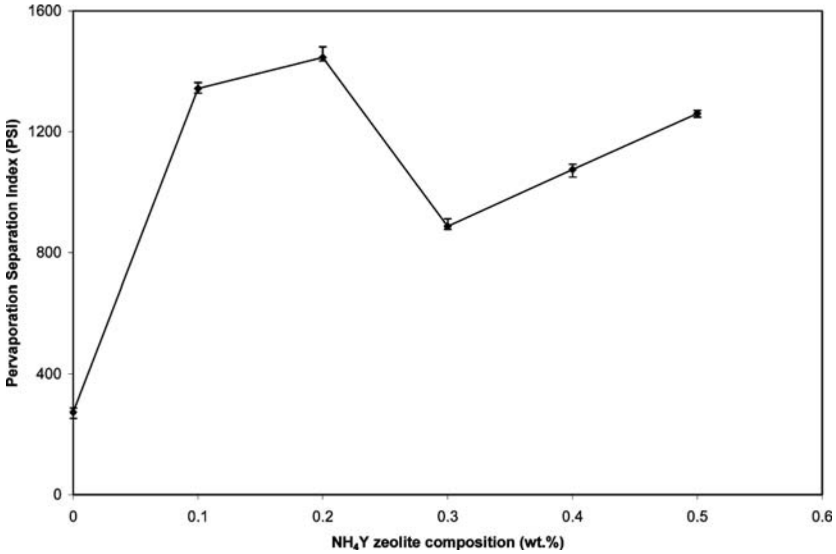


Figure 8. Effect of NH_4Y zeolite loading on pervaporation separation index at 10 wt.% of water in the feed.

NH_4Y zeolite loading gave the optimum performance in the pervaporation process for the dehydration of water-isopropanol mixture using the NH_4Y zeolite-filled membranes.

Comparison of Pervaporation Performance of Chitosan-Based Membranes

Table 1 summarizes the flux and separation characteristics of chitosan-based membranes for water-isopropanol mixtures. The Pervaporation Separation Index of zeolite NH_4Y-incorporated membranes is reasonably higher than those of composite and pure chitosan membranes (2, 3). However, the chitosan membranes cross-linked with sulfosuccinic acid (3) and glutaraldehyde (3) have significantly higher Pervaporation Separation Index even when the measured temperatures are high. This may be because of a lower thickness obtained in membrane preparation, which might have played a decisive role in achieving higher selectivity in addition to membrane property. Among the membranes developed in the present study, the 0.2 wt.% zeolite loaded membrane showed the permeation flux, which is comparable to the pure chitosan membrane (3). However, the permeation flux is lower compared to other composite membranes. Based on the comparison, it clearly reveals that the mass of zeolite loaded and membrane thickness play a key role in achieving higher permeation flux and separation factor. Hence,

Table 1. Comparative study of pervaporation performance of chitosan-based membranes reported in the literature for water-isopropanol mixtures

Membrane	Thickness (μm)	Temperature (°C)	Water in feed (wt%)	Flux (g/m^2h)	PSI	Ref.
Chitosan cross-linked with SSA	20	40	20	105	∞	5
Chitosan	40	30	5	32	422	5
Chitosan/TDI	50	30	8.4	79	472	2
Chitosan–polysulfone composite	NR	30	5	240	807	5
Chitosan cross-linked with HMDI	NR	30	5	82	1964	5
Chitosan–polysulfone composite cross-linked with GA	10	50	5	446	5912	5
Chitosan–NaY zeolite (10 wt% NaY zeolite)	40	30	5	48	729	5
Chitosan	30	30	10	30	273	#
Chitosan–NH_4Y zeolite (0.2 wt% NH_4Y zeolite)	30	30	10	39	1446	#

SSA: sulfosuccinic acid; GA: glutaraldehyde; HMDI: hexamethylene diisocynate; TDI: toluene-2,4 diisocyanate; NR: not reported in the literature; #: present work.

in the future we intend to prepare ultra-thin membranes by adopting a suitable method to improve both the permeation flux and the separation factor.

Effects of NH_4Y Zeolite Loading on Diffusion Coefficient

Diffusion data for molecules that are used in zeolite membrane pervaporation can improve understanding of transport mechanisms involved in the pervaporation and allow predictions of fluxes and selectivities. Permeation measurements on zeolite membranes have been used to indirectly infer multicomponent diffusion behavior (18).

Mass transport of liquid molecules in pervaporation experiments is generally explained by the solution-diffusion mechanism, which occurs in three steps: sorption, diffusion, and evaporation (3). Thus, the permeation rates and selectivity are governed by sorption and diffusion of each component through the membranes. The diffusion coefficient is used to describe the sorption and diffusion behavior of the system. Using the data of pervaporation experiment, the diffusion coefficient, D_i is calculated from the equation below (3):

$$J_i = \frac{D_i}{\delta} c_i \tag{9}$$

The diffusion coefficient is assumed to be constant across the effective membrane thickness, δ. c_i is the concentration of water or isopropanol in the feed after the pervaporation and J_i is the permeation flux of water or isopropanol. The computed values of diffusion coefficient are tabulated in the Table 2.

Table 2 shows that the values of diffusion coefficient calculated for water were always 100 to 1000 times higher than the values of diffusion coefficient calculated for isopropanol. This indicates that the membranes were highly hydrophilic and highly selective towards water. The increase of the NH_4Y

Table 2. Diffusion coefficient of water and isopropanol for the membranes at different NH_4Y zeolite loading calculated at 30°C with 10 wt.% of water in feed

Wt.% of NH_4Y zeolite	D_w ($\times 10^{12}$ m^2/s)	D_{IPA} ($\times 10^{14}$ m^2/s)
0.0	1.10	14.31a
0.1	2.15	7.70
0.2	2.19	7.60
0.3	3.46	42.11
0.4	3.39	29.75
0.5	3.87	34.49

zeolite loading in the chitosan membrane also increased the values of diffusion coefficient for water. This proves that the increase of NH_4Y zeolite loading lead to the increase of hydrophilic properties of the membranes. However, there was a considerable decrease in the diffusion coefficients of isopropanol for the zeolite loading of 0 wt.% to 0.2 wt.%. At this region, the membranes had low isopropanol permeation flux with high separation factor as shown in Fig. 6 and Fig. 7 respectively. This indicates that the NH_4Y zeolite-filled chitosan membranes at 0.1 wt.% and 0.2wt.% had the highest water selectivity, stability, and crystallinity which strongly resists the swelling effect. Nevertheless, the diffusion coefficients of isopropanol increased at the NH_4Y zeolite loading of 0.3 wt.% to 0.5 wt.%. This proves that at high NH_4Y zeolite loading, the swelling process which resulted some tiny cracks at the NH_4Y zeolite-chitosan interface allowed more isopropanol molecules to diffuse through the membrane (8).

CONCLUSIONS

Modification of the homogeneous chitosan membrane using NH_4Y zeolite caused non-homogeneous dispersion of NH_4Y zeolite crystals due to the precipitation of NH_4Y zeolite at the bottom of the membrane. In addition, the presence of NH_4Y zeolite in the chitosan membrane resulted in membrane swelling due to its hydrophilic properties. The degree of swelling increased with the increase of NH_4Y zeolite loading. The sorption selectivity decreased with the increase of NH_4Y zeolite loading although the water sorbed by the membrane was always higher than the isopropanol.

However, the presence of NH_4Y zeolite in the chitosan membrane had improved the overall performance of the membrane in separating water-isopropanol mixture using the pervaporation process. The NH_4Y zeolite was able to improve both tensile strength and percentage of strain at maximum of the chitosan membrane. 0.2 wt.% and 0.4 wt.% of NH_4Y zeolite loading had the highest tensile strength and strain at maximum respectively. This shows an improvement of 1.1 times and 1.5 times higher for tensile strength and strain at maximum respectively compared to the homogeneous chitosan membrane.

The increase of NH_4Y zeolite loading in the chitosan membrane resulted in increases of total, water and isopropanol permeation fluxes as well as the separation factor. The Pervaporation Separation Index shows that 0.2 wt.% of NH_4Y zeolite-filled membrane gave the optimum performance in the pervaporation process. Using 0.2 wt.% of NH_4Y zeolite-filled membrane, the total flux, separation factor, and Pervaporation Separation Index achieved an increase of 1.3, 3.8, and 5.3 times higher than the homogeneous chitosan membrane respectively.

The diffusion study proves that the modification of homogeneous chitosan membrane using NH_4Y zeolite increased the water selectivity and its hydrophilic properties thus giving high separation factor. However, at high loading of NH_4Y zeolite loading, the water selectivity decreased due to its swelling effect.

NOMENCLATURE

A	Membrane area
C_{IPA}	Weight fraction of permeate isopropanol in membranes at equilibrium sorption
C_w	Weight fraction of permeate water in membranes at equilibrium Sorption
c_i	Concentration of component *i* in the feed stream after pervaporation
D_i	Diffusion coefficient
J_t	Total permeation flux
J_i	Permeation flux of component *i*
Δt	Permeation time
W	Weight of permeate
w_f	Final (swollen) weight of the membrane
w_i	Initial (dry) weight of the membrane
X_w	Weight fraction of feed water in membranes at equilibrium sorption
X_{IPA}	Weight fraction of feed isopropanol in membranes at equilibrium sorption
x_w	Feed phase weight concentration of water
x_{IPA}	Feed phase weight concentration of isopropanol
y_w	Permeate phase weight concentration of water
y_{IPA}	Permeate phase weight concentration of isopropanol
$\alpha_{sorption\ w/IPA}$	Sorption selectivity
$\alpha_{w/IPA}$	Separation factor
δ	Membrane thickness

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