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A novel approach to fabricate zeolite membranes for pervaporation processes

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The conventional methods used in preparing zeolite membranes, such as the secondary growth and *in situ* crystallization methods, involve long and complex procedures that require the preparation of the zeolite aluminosilicate gel prior to the fabrication process and often result in membranes which contain pin holes. Here we report a simple, cheap, and less time-consuming technique to fabricate zeolite A and mordenite membranes on a porous stainless steel support. In addition, the technique makes it possible to fabricate types of zeolite membranes that have been previously difficult to synthesise. A clinoptilolite membrane was fabricated to demonstrate the ability to manufacture a zeolite membrane from an existing crystalline zeolite (natural or synthetic). All three membranes were subjected to separation tests, (ethanol dehydration, ethanol–cyclohexane and phenol removal from water) to demonstrate the efficacy of membrane synthesis. The fluxes obtained and separation factors which were achieved are comparable with literature values but as with most zeolite membranes there is a trade-off between high flux and separation.

Introduction

Membrane technology has attracted considerable interest among many research groups due to the separation performance, the simplicity of the concept and the low energy consumption.[1–4] In recent years, there have been extensive efforts in the petrochemical industries to develop more economical separation methods to replace conventional methods. This is due to the consumption of large quantities of energy used in, for example distillation processes.[5] Pervaporation has been recognized as a potentially viable low energy alternative for separation. Also, pervaporation overcomes numerous other separation difficulties, including the separation of mixtures in which components have similar boiling points (especially in the case of isomeric components), organic removal from water[6] as well as the separation of azeotropes. Many types of membranes have been examined, including polymeric and ceramic membranes. Researchers have studied zeolite membranes due to their uniform microstructures and their molecular sieving properties.[7,8] The ability of zeolites to handle the extreme operating conditions required by some separation technologies is well known. Zeolite properties are due to their three-dimensional, aluminosilicate, crystalline framework structures that form uniformly-sized pores of molecular dimensions.[9] The development of an anisotropic zeolite membrane that consisted of a very thin layer of zeolite supported on a thicker, porous structure made a major contribution to enhance zeolite membrane performance in terms of fluxes[10,11] since the pore dimensions of zeolites mean that thick, self-supporting, membranes have prohibitively low fluxes. However, zeolite membranes are usually fabricated using a hydrothermal, crystallization process that requires growth from a synthesis gel.[12,13] This initial step, due to its complexity is time consuming, and energy intensive and, is a significant obstacle to the developing and testing of many types of zeolite membranes.[2] Therefore, a technique for synthesising a given type of zeolite membrane without first having to synthesise the zeolite would be a valuable breakthrough from both an economic and technical perspective. In this paper, a simple, low cost and straightforward technique for the production of different types of zeolite membranes is presented. This technique was achieved by incorporating a low cost carbon precursor (sucrose solution) into the fabrication process. The objective of involving a carbon precursor in the synthesis process was to heal and block defects, similar approaches have been applied to synthesise nanoporous carbon membranes using furfuryl alcohols and polyimides.[14–16] The results presented in this paper indicate that effective membranes were synthesised for hydrophilic zeolite membranes, such as zeolite A, mordenite, and, for the first time, clinoptilolite. This allows the production of zeolite membranes but avoids hydrothermal synthesis replacing it with the direct use of pre-formed synthetic or natural zeolites.

Experimental

Membrane preparation

The overriding aim of the work is to produce a simple and repeatable method of producing zeolite membranes. The method which has been developed can be summarised as

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firstly, the introduction of a powdered zeolite to the surface of a porous support in the form of a slurry in water. Secondly, the zeolite on the support is dried prior to the addition of a filler which has the aim of blocking the inter-crystalline voids between the crystals of zeolite. This filler is a sucrose solution which is pulled through the zeolite/support by applying a vacuum. Finally, the filler is carbonised to produce a carbon structure which is impermeable, hence the permeation through the membrane takes place through the pores of the zeolite.

Sucrose (Fisher Scientific) was used as a filler because it is simple to prepare and does not require any pre-treatment. Moreover, sucrose has the advantage of being a natural resource which is readily available. The support metal used in fabricating the zeolite membranes were circular, porous, stainless-steel disks with a diameter of 20 mm, thickness of 1.5 mm and porosity of 0.5 μm. These disks were obtained from Aegis Advanced Materials Ltd., UK. The porous metal disks were washed by soaking them in a mild detergent solution (Dish-bac liquid detergent), after which they were sonicated in deionised water for 3 hours at room temperature. The washed metals were left to dry overnight at room temperature.

In this study, zeolite A (BDH Chemicals Ltd), mordenite (Zeolyst International) and clinoptilolite (Holistic Valley) were used. The zeolite paste was prepared by mixing deionised water and the synthetic or natural zeolite source in the ratio of 1 : 2 by weight. Then, 0.3 g of the prepared paste was applied to the porous, stainless-steel supports and spread over the surface of the disk using a small paint brush. After the paste was applied to the metal disks, it was allowed to dry for one hour at room temperature, after which a sucrose solution was added. The sucrose solution was prepared with different concentrations of sucrose–water ratios by weight (0.5 : 1, 0.7 : 1, 1 : 1, and 3 : 1) and 0.5 g of the solution was applied over the zeolite/support under suction through the membrane, the low viscosity solutions were added drop wise with a pipette, the higher viscosity (3 : 1 ratio) were spread over the surface with a spatula. After addition of the sucrose solution, the membranes were placed in a furnace and the sucrose carbonized by heating under flowing nitrogen. The temperature was raised at a rate of 5 °C min^{-1} up to 650 °C where it was held for a dwell period of 240 minutes prior to cooling to room temperature.

After carbonizing the sucrose the excess carbon/zeolite which was loosely bound to the surface of the disk was gently removed using treatment with a cloth as a mild abrasive. The remaining zeolite and carbon appears to be predominantly within the pores of the stainless steel support. Clearly the thickness of the membrane is difficult to measure when it is internal to the support however based on the weights of the disks before and after synthesis, the mass of carbon/zeolite can be measured. The results are shown in Table 1 for Zeolite A and the final weights for all 3 zeolites are summarised in Table 2. The change in mass is not an accurate measure of how much of the carbon/zeolite composite inside the support is 'active' in terms of separation since some loose material may be retained within the stainless steel support. Hence, the fluxes and separation factors for more than one membrane of each zeolite are reported to demonstrate consistency of performance.

In order to ensure the integrity of the zeolite structure during the pyrolysis conditions, all three zeolite were treated using the heat treatment described and the XRD pattern of the initial and final materials were compared to ensure that the thermal treatment did not affect the crystallinity. The XRD patterns were obtained using a Miniflex (Rigaku, X-ray analytical instrumentation) diffractometer.

Membrane testing

The pervaporation rig used to evaluate the performance of the membranes consisted of six major elements shown in Fig. 2 a feed tank, liquid pump, heater, membrane compartment, permeate traps, and a vacuum pump. The feed mixture was placed in a container to prevent heat loss and to avoid evaporation, the equipment also allowed for heating of the feed when required. The design of the membrane compartment used in this study contained two hemispherical glass sections. The prepared membranes were fixed in the membrane compartment by fixing on to a nonporous metal washer using an epoxy adhesive. Then, membranes attached to the washers were fitted between two Teflon rings and sealed with vacuum grease between the hemispherical glass sections. Finally the hemispherical glass compartments were clamped together. The pervaporation process was conducted with varying feed compositions with total volume of 200 ml, at atmospheric pressure on the feed side. On the permeate side, the pressure was set to 8 Pa, and the permeate (vapour phase) was collected by condensing it with liquid nitrogen traps. The performance of each membrane used in the process described above was determined in terms of separation factor and total flux.

-The separation factor ($\alpha_{i,j}$) was calculated from:

$$\alpha_{i,j} = \frac{W_{\mathrm{P},i} W_{\mathrm{F},j}}{W_{\mathrm{P},j} W_{\mathrm{F},i}}$$

Table 1 Zeolite A membrane weight during preparation (1 : 1 water sucrose ratio)[a]

	Weight (grams)	
Zeolite A membrane	Sample 1	Sample 2
S.S. disc	3.0464	3.0462
S.S. disc + zeolite paste	3.3478	3.3470
S.S. disc + zeolite paste + sucrose	3.8352	3.8481
Membrane after pyrolysis	3.0611	3.0580
Mass of carbon plus zeolite	0.0147	0.0118

[a] S.S. = stainless steel.

Table 2 Mass of carbon plus zeolite for each membrane (1 : 1 water sucrose ratio)

	Weight (grams)	
Membrane	Sample 1	Sample 2
Zeolite A plus carbon	0.0147	0.0118
Mordenite plus carbon	0.0046	0.0031
Clinoptilolite plus carbon	0.0046	0.002

where, (W_i) and (W_j) are the weight composition of the binary components of the mixture and P and F indicate the weight composition of permeate and feed, respectively.

-Total permeate flux (F) was calculated from:

$$F = \frac{m_s}{A\Delta t}$$

where (m_s) is the weight of the collected permeate sample, (A) is membrane surface area and (Δt) experiment duration.

Results and discussion

Membrane synthesis

The XRD patterns shown in Fig. 1 demonstrate that the zeolite crystal structure is unchanged by thermal treatment at 650 °C and as such we can assume that the zeolite in the membranes is similarly unaffected. While there is some difference in the total weight of the carbon plus zeolite in the synthesised membranes, the repeat samples in Tables 3–5 show that the membranes themselves are very similar in terms of performance and hence 'effective' thickness.

Ethanol dehydration

Zeolite A membranes were evaluated using a range of ethanol–water mixtures, 4, 6, and 20 wt% water. The results of the separation experiments, which are presented in Table 3, show that the membranes had comparable separation and fluxes when compared to other similar membranes in the literature at similar temperatures[17,18] The effect of the carbon precursor solution was tested and evaluated at three different concentrations, as shown in Table 3. The results indicate that the concentration of the carbon precursor solution has a significant influence on the performance of the membranes in terms of flux and separation factor. As with most zeolite membranes, there is a trade-off between flux and separation, as the concentration of sucrose increases, the flux decreases and the separation factor increases. This occurs up to a limit where the viscosity of the sucrose solution does not facilitate the formation of thin, coherent membranes. The membrane prepared with the highly concentrated sucrose solution (3 : 1) did not show a performance that was consistent with those of the other three membranes. This indicated the difficulty of passing the concentrated viscous sucrose solution through the narrow pinholes to reach the bottom of the zeolite paste layer on the support metal. The SEM analysis (top view) presented in Fig. 3 shows that the zeolite carbon composite made with less than 3 : 1 sucrose water ratio is within the pores of the stainless steel support but is hence difficult to image by SEM.

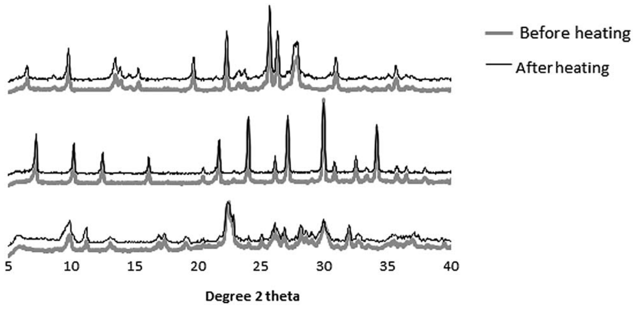

Fig. 1 Comparison of zeolite a, mordenite and clinoptilolite before and after applying pyrolysis conditions.

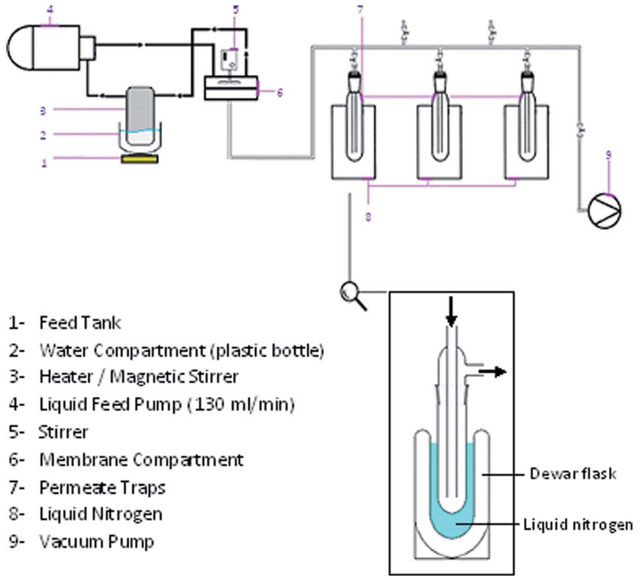

Fig. 2 Schematic diagram of pervaporation unit with membrane module.

The performances of membranes Z-S.1 to Z-S.4 were evaluated using three different feed compositions of ethanol (96, 94, and 80 wt% in water), and the influence on both of the permeate fluxes and separation factors is illustrated in Fig. 4 and 5, respectively. A pure sucrose membrane was prepared (without zeolite) to assess its permeability. This sucrose membrane did not allow any component to permeate over a 12 hour period which demonstrates the efficacy of carbonised sucrose as a filler. Therefore, it can be concluded that the concentration of the carbon precursor solution is related to the separation selectivity and inversely related to the overall flux. The choice of the concentration of the solution is of great importance and should be made based on the desired separation behaviour of a given membrane.

Although the results showed an improvement of the zeolite membrane in terms of performance and the simplicity of preparation, the separation factors of the membranes prepared in this work were lower than those in the literature for pure zeolite A membranes on stainless steel supports.[19] Holmes and co-workers achieved a separation factor of 198.75 at 25 °C with a feed of 80 wt% ethanol. However, in this work the advantages are of a shorter preparation time and higher fluxes. Holmes and co-workers fabricated membranes using the secondary growth method (SGM) after a crystallization process of four days, resulting in a flux of 58 (g m^{-2} h^{-1}), whereas the novel technique used in this study was conducted for less than one day (10 h). A comparison was conducted at 25 °C with a feed composition of 80 wt% ethanol, and the results of the comparison are shown in Fig. 6. This again illustrates the trade-off between flux and separation.

Table 3 Evaluation of carbon–zeolite membranes using different sucrose solution concentrations (as a carbon precursor) after coating the porous support with zeolite A paste, at different feed compositions of ethanol–water mixture (Z-S.3a is a different membrane manufactured in the same fashion as Z-S.3, to show reproducibility)[a]

Membrane	Sucrose–water ratio (weight)	Feed (wt%)		Permeate (wt%)		Permeate flux (g m^{-2} h^{-1})	Separation factor
		Water	EtOH	Water	EtOH		
Z-S.1	0.5 : 1	4	96	46.68	53.32	1072.75	21.59
		6	94	58.23	41.77	1086.86	20.89
		20	80	79.67	20.33	1151.79	17.52
Z-S.2	0.7 : 1	4	96	51.04	48.96	1013.46	25.71
		6	94	59.01	40.99	1026.87	21.58
		20	80	80.64	19.36	1065.69	18.62
Z-S.3	1 : 1	4	96	65.67	34.33	931.59	47.18
		6	94	73.68	26.32	960.53	41.97
		20	80	82.23	17.77	981.70	20.69
Z-S.3a	1 : 1	4	96	66.95	33.05	917.48	49.96
Z-S.4	3 : 1	4	96	60.68	39.32	47.05	38.06
		6	94	68.22	31.78	52.93	32.18
		20	80	79.91	20.09	57.63	17.78

[a] EtOH: ethanol.

Using a mordenite membrane, made with 1 : 1 sucrose to water ratio, a similar experiment was conducted in which ethanol–water mixtures were separated. The experiment was again conducted at 20 °C with ethanol–water mixtures that consisted of 2 and 6 and 10 wt% water. The results of the separation are shown in Table 4.

The SEM images in Fig. 7 show a top view of the mordenite carbon composite on the porous stainless steel disk. Unlike the zeolite A, the image shows that there is a significant quantity of zeolite apparent on the external surface of the disk.

The separation factor was high compared to the zeolite A membrane used in this study, reaching 634, and the fluxes were competitive (15 g m^{-2} h^{-1}).

The membranes of this current study had better separation factors than those reported in the literature by Navajas *et al.* using 10 wt% ethanol.[20] Although Navajas *et al.* achieved greater permeate fluxes (200 g m^{-2} h^{-1}), the method they used, *i.e.*, the secondary growth method (SGM), took a long time and required the preparation of the zeolite gel. Fig. 8 presents the outcomes of the current study and a comparison with those of Navajas and co-workers.

Separation of ethanol–cyclohexane mixture

To test the feasibility of fabricating a membrane with a zeolite that was acquired from natural sources, a clinoptilolite membrane was fabricated using the same technique. Clinoptilolite has pore dimension of approximately 0.6 nm, which makes it suitable for many separation applications. Moreover, the fabrication of this type of zeolite membrane has not been reported in the literature due to the complexity of its hydrothermal synthesis.

After clinoptilolite membranes were prepared, they were evaluated using ethanol–cyclohexane mixtures. This mixture was used to take advantage of the difference between the properties of the mixture in terms of polarity and the molecular dimensions. Methanol and ethanol have lower polarity than water, but, compared to other organic solvents, they can be considered as polar molecules.[21] Ethanol was chosen over methanol because it has lower polarity than methanol and is miscible in cyclohexane. The ethanol–cyclohexane mixture offers the advantage of different size molecules, *i.e.*, the kinetic diameters of ethanol and cyclohexane are 0.43 and 0.60 nm, respectively. The results of the experiments showed that the first

Table 4 Evaluation of mordenite membranes using sucrose as a carbon precursor at different feed compositions of water–ethanol mixture. The repeat sample is a different membrane manufactured in the same fashion to show reproducibility[a]

Membrane	Sucrose–water ratio (weight)	Feed (wt%)		Permeate (wt%)		Permeate flux (g m^{-2} h^{-1})	Separation factor
		Water	EtOH	Water	EtOH		
Mordenite	1 : 1	10	90	97.72	2.28	15.70	388.89
		6	94	97.69	2.31	15.23	633.97
		2	98	88.86	11.14	14.53	623.057
Mordenite (repeat)	1 : 1	10	90	97.53	2.47	15.23	358.27

[a] EtOH: ethanol.

Table 5 Evaluation of carbon–zeolite membranes using clinoptilolite and sucrose as a carbon precursor with different feed compositions at room temperature, 40 and 60 °C[a]

Membrane	Temperature °C	Sucrose–water ratio (weight)	Feed (wt%)		Permeate (wt%)		Permeate flux (g m^{-2} h^{-1})	Separation factor
			EtOH	Chx	EtOH	Chx		
Clinop.1	25	0.5 : 1	5	95	Leaking			
Clinop.1	25	1 : 1	5	95	32.59	67.41	183.49	9.07
			6	90	47.56	52.44	197.61	8.06
			20	80	49.64	50.36	239.96	3.89
Clinop.2	25	1 : 1	5	95	36.68	63.32	225.84	10.87
			6	90	50.67	49.33	228.66	9.13
			20	80	51.03	48.97	238.54	4.12
Clinop.2	40	1 : 1	5	95	35.89	64.11	268.19	10.50
			6	90	47.67	52.33	296.42	8.09
			20	80	52.67	47.33	409.34	4.39
Clinop.2	60	1 : 1	5	95	33.45	66.55	437.57	9.43
			6	90	49.93	50.07	479.91	8.86
			20	80	51.12	48.88	536.37	4.13

[a] EtOH: ethanol; Chx: cyclohexane.

membrane, which was prepared with a concentration of sucrose to water of 0.5 : 1 by weight, did not provide any separation therefore this membrane was subjected to further treatment using the carbon precursor at the higher concentration of 1 : 1. The performance of the membrane after treatment is listed in Table 5, and it reached a separation factor of 13.15 with fluxes around 200 g m^{-2} h^{-1}, this result demonstrated the potential benefit of subjecting a leaking membrane to sucrose 'healing', by applying a suitable concentration of the carbon-precursor solution (sucrose). For comparison, the fabrication of a clinoptilolite membrane was repeated, starting with a sucrose-solution concentration of (1 : 1). An SEM image of the membrane, is shown in Fig. 9. The clinoptilolite morphology can be observed in the SEM images, and again, as in mordenite, there appears to be significant crystal deposit on the external surface of the disk. The performance of the second membrane was slightly better than that of the first one in terms of the overall fluxes, and this was due to the repeated treatment by the sucrose solution in the first membrane. In order to understand the effect of temperature, these membranes were evaluated further at different operating temperatures, *i.e.*, 40 and 60 °C, on the feed side of the membrane cell. As indicated in Table 5, the temperature influences the overall fluxes but it did not make a noticeable difference to the separation factors.

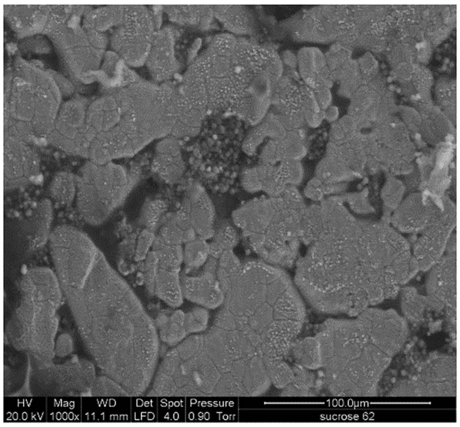

Fig. 3 Top view of zeolite a sucrose composite on porous stainless steel disk showing the zeolite to be predominantly internal to the support.

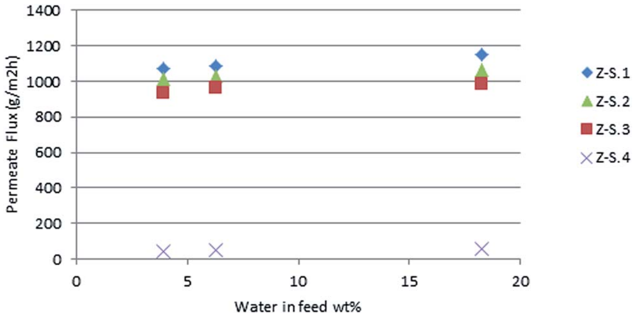

Fig. 4 Illustration of the overall flux dependence on the feed composition using zeolite membranes.

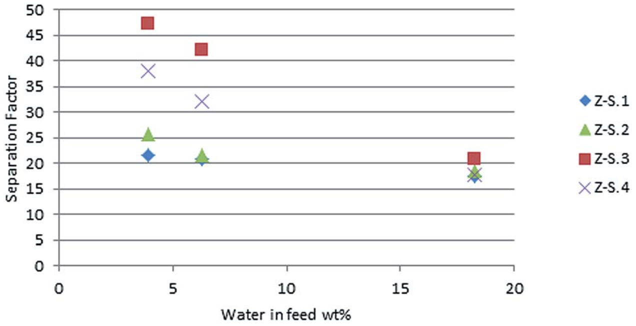

Fig. 5 Illustration of the separation factor dependence on the feed composition using zeolite membranes post-treated with sucrose.

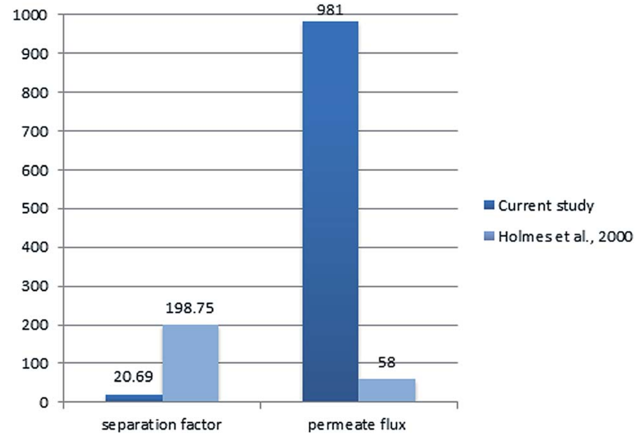

Fig. 6 Comparison of the performances of the membranes prepared by Holmes *et al.* and those in the current study at 25 °C using a mixture that contained 80 wt% ethanol.

Removal of phenol from water

Phenol has been used as a raw material for the production of many chemicals including caprolactam, which is consumed in the production of nylon fiber. Therefore, a large quantity of aqueous waste streams containing phenols are discharged by numerous industries. Consequently, removing phenol from water is of great importance in the wastewater treatment industry.[22,23]

Results are presented for the testing and evaluation of the zeolite A and clinoptilolite membranes with the phenol–water mixture. Both of these membranes had the appropriate polarity and pore-dimensions to carry out this separation. Zeolite A and clinoptilolite are classified as hydrophilic zeolites due to the Si/Al ratios in these membranes. Their silicon to aluminium ratios are 1.1 and 5.25, respectively based on the results of EDAX analysis.

The kinetic diameter of phenol molecules (0.66 nm) is larger than the pores of both zeolite A (0.41 nm) and clinoptilolite (0.60 nm). Since the solubility of phenol in water is 8.3 g/100 ml at 20 °C, a solution of 5 wt% phenol was prepared by mixing 5 g of phenol (obtained from Sigma Aldrich) with 95 g of deionised water at room temperature; this solution was fed to the membrane cell using zeolite A and clinoptilolite membranes. In general, the observed performances of these membranes showed that water was highly preferred over phenol for both of these membranes, as shown in Table 6. However, zeolite A had a better separation factor than clinoptilolite, but the fluxes in case of clinoptilolite were much greater. The separation behaviour of clinoptilolite at different temperatures is presented in Table 7

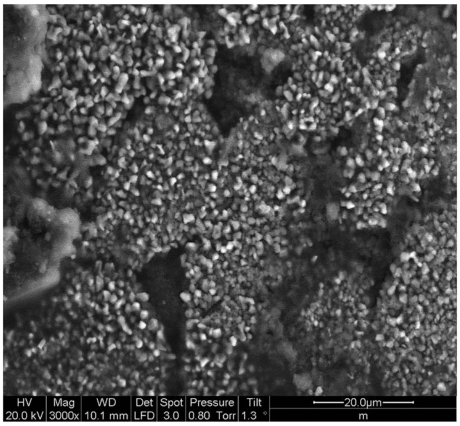

Fig. 7 SEM image of the top view of carbon–mordenite composite layer in the pores of the stainless steel disk showing some zeolite distribution on the surface.

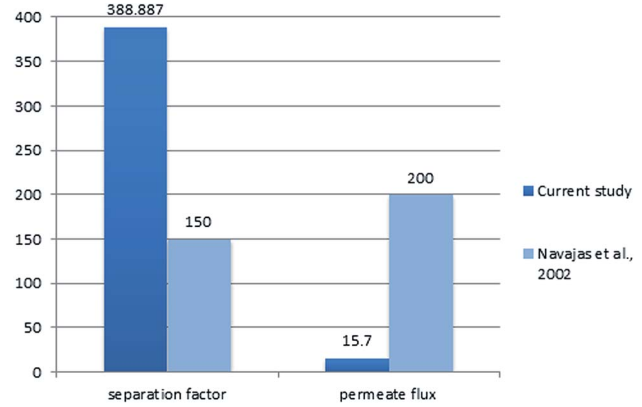

Fig. 8 Comparison of the performances of mordenite membranes prepared by Navajas *et al.*[19] and those prepared in the current study.

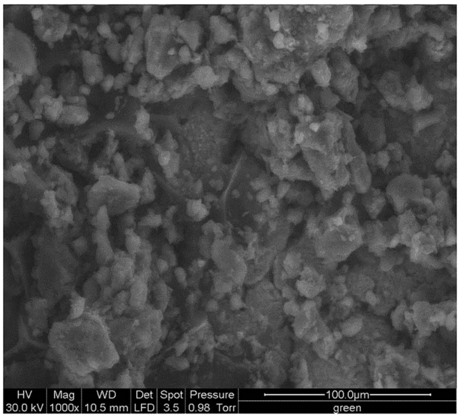

Fig. 9 SEM image of carbon–zeolite clinoptilolite composite layer (1 : 1, sucrose : water).

Table 6 Evaluation of zeolite A and clinoptilolite membranes using sucrose solution of (1 : 1) concentration for phenol–water separation at 25 °C

Membrane	Feed (wt%)		Permeate (wt%)		Permeate flux (g m^{-2} h^{-1})	Separation factor
	Phenol	Water	Phenol	Water		
Zeolite A	5	95	0.34	99.66	70.57	16.51
Clinoptilolite-1	5	95	0.37	99.63	303.47	15.16

Table 7 Evaluation of clinoptilolite membrane using sucrose solution of (1 : 1) concentration for phenol–water separation at different feed temperatures

	Feed (wt%)		Permeate (wt%)			
T (°C)	Phenol	Water	Phenol	Water	Permeate flux ($g\ m^{-2}\ h^{-1}$)	Separation factor
25	5	95	0.36	99.64	299.77	15.76
40	5	95	0.41	99.59	311.41	13.68
60	5	95	0.39	99.61	319.35	14.38

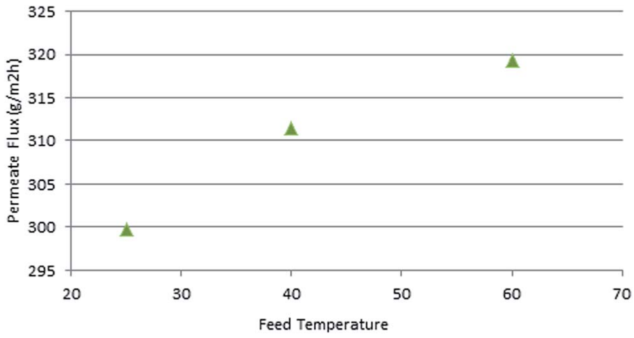

Fig. 10 Illustration of temperature effect on permeate fluxes using clinoptilolite-2 membrane.

and the data indicate that they all had similar separation performances while the overall fluxes increased as the temperature increased, as shown in Fig. 10.

The results achieved in this work were compared with those in the literature provided by Pradhan *et al.*[22] in a study in which they separated phenol–water mixtures using two different types of membranes, *i.e.*, polyimide and polyimide with lithium chloride (LiCl) at 40 °C. Fig. 11 and Table 8 show that this current study had better performance in terms of separation factor and total fluxes.

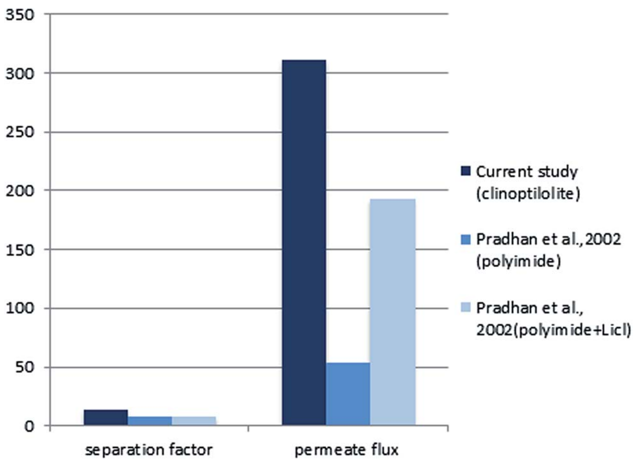

Fig. 11 Comparison of the performances of membranes prepared in this study and those prepared by Pradhan *et al.*

Table 8 Comparison of the performances of membranes prepared in this study and those prepared by Pradhan *et al.*

			Feed (wt%)			
Membrane	Reference	T (°C)	Water	Phenol	Permeate flux ($g\ m^{-2}\ h^{-1}$)	Separation factor
Polyimide	Pradhan *et al.*	40	96	4	54	8.6
Polyimide with LiCl	Pradhan *et al.*	40	96	4	193	7.61
Clinoptilolite	Current study	40	95	5	311.41	13.68

Table 9 Evaluation of carbon–zeolite A membrane using sucrose solution of (1 : 1) concentration for ethanol dehydration at 25 °C, and feed composition of 96% of ethanol[a]

	Feed (wt%)		Permeate (wt%)			
Intervals	Water	EtOH	Water	EtOH	Permeate flux ($g\ m^{-2}\ h^{-1}$)	Separation factor
Week 1	4	96	68.53	31.47	1009.23	53.71
Week 2	4	96	69.02	30.98	988.06	54.95
Week 5	4	96	68.88	31.12	1037.46	54.59
Week 8	4	96	68.22	30.78	1016.29	55.46

[a] EtOH: ethanol.

Evaluation of quality and durability

The durability of the carbon–zeolite membranes that were synthesized was tested and assessed. The carbon–zeolite A membrane was tested after 1, 2, 5 and 8 weeks for separation of an ethanol–water mixture using the pervaporation process for 30 min at 25 °C and at a feed concentration of 96% of ethanol. During the period between tests the membrane was maintained under a constant composition ethanol–water mixture The results shown in Table 9 indicate that the method of fabricating zeolite A introduced in this study yielded a membrane that had stable performance up to two months. Since the other types of zeolite membranes followed the same fabrication procedure, they are expected to have similar durabilities as the solvent does not appear to degrade the carbon or the zeolite structure or the interface between the two.

Conclusion

The preparation of zeolite membranes has been successfully achieved using this simple, healing, technique. Three types of zeolite membranes were fabricated successfully using this method, *i.e.*, zeolite A, mordenite and clinoptilolite. The successful synthesis of a clinoptilolite membrane demonstrated the feasibility of fabricating a membrane that had not been reported before due to the complexity of preparing clinoptilolite by hydrothermal synthesis.

Zeolite A and mordenite membranes were tested with ethanol–water mixtures, and they had performances that were competitive with those presented in the literature *i.e.*, Holmes *et al.*[19] and Navajas *et al.*[20]

Clinoptilolite membranes prepared in this study had greater fluxes and greater selectivity than the polyimide membranes prepared by Pradhan *et al.*[22]

It is clear that there is a trade-off between flux and separation factor and this could be optimised for a given system by altering the quantity of sucrose used. The separation factors achieved clearly demonstrate the efficacy of the carbon treatment process.

In summary, the ability to fabricate any type of zeolite membrane using the novel method presented in this work is step forward in the zeolite membranes synthesis landscape, since it was achieved with a simple procedure and requires a much shorter fabrication time than the conventional method.

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