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Optimization of synthesis conditions for preparation of ceramic (A-type zeolite) membranes in dehydration of ethylene glycol

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Abstract

High quality ceramic (A-type zeolite) membranes were synthesized on α-Al_2O_3 as substrate with the aid of sub-micro zeolite powder as seeds. The zeolite membranes and seeds were synthesized via hydrothermal process. The influences of synthesis conditions including synthesis time and synthesis temperature on structure and permeation properties of the A-type zeolite membranes were investigated. The synthesized membranes were characterized using X-ray diffraction (XRD) and scanning electron microscope (SEM). Sub-micro seeds of A-type zeolite with high crystallinity and crystallite size ranging from 150 to 350 nm were synthesized successfully. The best A-type zeolite membranes with the aid of sub-micro seeds were obtained for synthesis time of 2 h and at synthesis temperature of 100 °C, exhibiting permeation flux of 4.03 kg m^{-2} h^{-1} and separation factor of more than 10,000 for dehydration of the 10:90 (wt%) water/ethylene glycol (EG) mixture at 70 °C. Moreover, the separation performance of membranes for dehydration of 10:90 (wt%) water/ethanol at 70 °C showed total flux and separation factor of 11.14 kg m^{-2} h^{-1} and 10,000, respectively. It was observed that synthesis of high quality A-type zeolite membranes need optimization of time and temperature during synthesis of zeolite layers.

Keywords: Ceramic membrane; LTA zeolite; Hydrothermal; Pervaporation; Ethylene glycol

1. Introduction

Great characteristics of ceramic membranes such as higher mechanical strength, chemical and thermal stability, compared with other inorganic and polymeric membranes have made broad applications in gas separation, wastewater treatment, pervaporation, membrane reactors, and chemical sensors. Zeolite membranes have attracted many attentions as well-defined crystalline materials for separations and reactions [1–10]. Different types of zeolite membranes, including LTA, MFI, X, Y, W, MOR, SOD, and T have been successfully prepared [11–19].

Compared to conventional membrane separation processes, a few zeolite membranes have been industrialized. The first industrial separation process using A-type zeolite membranes were installed by Mitsui Engineering and Shipbuilding Co. Ltd for dehydration of alcohols. However, installation of zeolite membranes has not been developed significantly due to two main problems: (1) economical feasibility and (2) incomplete development of synthesis procedure including poor reproducibility [20–22].

Some solutions have been suggested for development of zeolite membranes synthesis to overcome the mentioned problems. To overcome the economical problem, preparation of high flux membranes would reduce both costs of membranes and modules fabrication [21]. The poor quality of A-type zeolite membranes could be due to improper synthesis procedures. To improve the quality of zeolite membranes and control the zeolite layers growth, seeding technique is currently used. In this method, substrate is covered by zeolite powder (seeds) before membrane synthesis. During the synthesis of A-type zeolite membranes, seeds accelerate the crystallization rate of A-type zeolite on the support surface. However, in most studies, commercial A-type zeolite powders have been utilized as the nucleation

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seeds. The problem is that, the commercial A-type zeolite powders have large particle size and broad particle size distribution. This results in the formation of deficient membranes. It is difficult to form a high-quality zeolite membrane by zeolite seeds with a broad particle size distribution [23–25].

Moreover, synthesis rate of zeolite layers is affected by the particle size of zeolite seeds. Smaller seeds accelerate zeolite membrane synthesis with faster crystallization rates. Therefore, when nano powders are used for seeding a substrate, a defect free zeolite layer (membrane) can be formed homogeneously [26,27]. Compared with commercial seeds, when nano seeds are used for membrane synthesis, increasing synthesis time increases membrane thickness which in turn reduces membrane permeation flux. With further increasing synthesis time, different zeolites may be formed. Thus, further optimization of synthesis conditions is necessary to prepare the best zeolite membranes.

One of the applications of A-type zeolite membranes is dehydration of alcohols. Dehydration of some alcohols such as ethanol, dimethylformamide, methanol, and isopropyl alcohol using A-type zeolite membranes have been widely studied [28–33]. Ethylene glycol (EG) is an important chemical material which is mostly used as nonvolatile antifreeze and coolant as well as intermediate in manufacture of polyesters [34].

Conventional synthesis of EG includes hydrolysis of ethylene oxide using excess water to improve EG yield. The product requires an extra dehydration process to obtain pure EG [35].

EG has very high boiling point (about 200 °C), and therefore is known as a heavy chemical. Removal of water from EG/water liquid mixtures ranks as the eighth most energy intensive distillation process in chemical industries [36]. Therefore, it is justified to use membrane technology for dehydration of EG.

A few studies have been performed regarding dehydration of EG using A-type zeolite membranes. Nik et al. studied performance of A-type zeolite membrane for dehydration of EG using pervaporation (PV). A flux of 0.94 kg m^{-2} h^{-1} and a separation factor of 1177 were achieved at 70 °C for a feed of 30 wt% water in EG solution [36].

Recently Yu et al. have synthesized zeolite A-type membranes on surface of porous tubular substrates of mullite. The membranes were used for dehydration of EG via PV. For a feed solution of 10 wt% water in EG at 120 °C, a flux of 1.83 kg m^{-2} h^{-1} and a separation factor more than 4000 were obtained [35]. However, their operation temperature is very high for dehydration of EG which is not favorable in PV.

The results showed that A-type zeolite membranes have great potential in dehydration of EG. Compared to other organic solvents, water flux in EG dehydration by A-type zeolite membrane is usually lower [36]. Therefore, more investigations on optimization of the synthesis procedure is of vital importance.

In this work, the uniform sub-micro size A-type zeolite powder was synthesized via hydrothermal technique. The sub-micro powder was then used as seeds for synthesis of A-type zeolite membranes. High-quality pure A-type zeolite membranes were synthesized via secondary growth method using sub-micro seeds and optimization of synthesis temperature and synthesis time was also performed. The prepared membranes were then characterized using X-ray diffraction (XRD) and scanning electron microscope (SEM). The membranes performance was finally investigated using dehydration of EG in PV to determine their permeation flux and separation factor.

2. Experimental

Sub-micro seeds and membranes of A-type zeolite were synthesized via hydrothermal method in a PTFE autoclave. The used reagents for synthesis of sub-micro seeds were aluminum isopropoxide (Merck, 97% C_9 H_{21} Al O_3) as source of Al and colloidal silica (Ludox, Aldrich 50% SiO_2) as source of Si. On the other hand, sodium aluminate (Riedel de Haen, 50–56% Al_2O_3, 40–45% Na_2O) and sodium silicate (Merck, 25.5–28.5% SiO_2, 7.5–8.5 Na_2O) were used for the membranes synthesis. Sodium hydroxide (Merck, 98% NaOH) and deionized water are also used for synthesis of both sub-micro seeds and membranes.

2.1. Synthesis of NaA sub-micro seeds

Composition of the synthesis gel for sub-micro A-type seeds was 1.0 Al_2O_3:2.7 SiO_2:5.85 Na_2O:150H_2O. The synthesis solution was prepared by mixing aluminate and silicate precursors. NaOH (7.838 g) was dissolved in 42.759 ml of deionized water. The solution was divided into two equal volumes and kept in polypropylene bottles. Aluminate solution was prepared by adding 6.946 g aluminum isopropoxide to one part of the solution. It was mixed at 60 °C until cleared. Silicate solution was prepared by mixing 4.546 g Ludox to another part of the NaOH solution at 60 °C until cleared. After cooling the two solutions down to 25 °C, the silicate solution was then poured into the aluminate solution and mixed until a thick homogenized gel was formed. The formed gel was then aged for 48 h at room temperature and then, the gel was heated at 60 °C for 11 h. After synthesis of the seeds, the sub-micro particles were recovered by washing with deionized water using filter papers (Whatman Grade 3), until pH of the seeds suspensions became close to 8–9.

2.2. Seeding of supports

Homemade porous α-alumina disks with thickness of 1.8 mm and diameter of 21 mm, water permeation of 190 kg m^{-2} h^{-1} bar and porosity of 43% were used as supports. The surface of the supports was polished with 1500 grit-sand papers, and cleaned with deionized water in an ultrasonic cleaner (Branson SB2200) several times to

remove the loose particles created during polishing. The NaA zeolite seeds were carefully selected. The selected seeds had small particle size and high crystallinity. Dip coating method was used for seeding the supports. A colloidal suspension was prepared by dispersing 1 g NaA zeolite seeds in 100 ml deionized water with ultrasonic treatment. The supports were immersed in the suspension for 30 s and then dried at 60 °C overnight.

2.3. Synthesis of A-type zeolite membranes

A-type zeolite membranes were prepared hydrothermally on the porous seeded supports. The synthesis solution was prepared by mixing aluminate and silicate solutions. NaOH (1.977 g) was dissolved in 41.583 ml of deionized water. The solution was divided into two equal volumes and kept in polypropylene bottles. Aluminate solution was prepared by adding 3.238 g sodium aluminate to one part of the NaOH solution. It was mixed until cleared. Silicate solution was prepared by adding 7.557 g sodium silicate to another part of the NaOH solution. Silicate solution was then poured into aluminate solution and mixed until a thick homogenized gel was formed. Composition of the homogeneous solution of A-type zeolite is represented by the following molar ratio: 1 Al_2O_3:2 SiO_2:3.4 Na_2O:155H_2O [26].

One side of the supports was covered by Teflon tape to prevent membrane growth on both sides of the supports. A Teflon holder was used to locate the seeded supports vertically in the autoclave during the synthesis of membranes. Schematic of the autoclave is presented in Fig. 1.

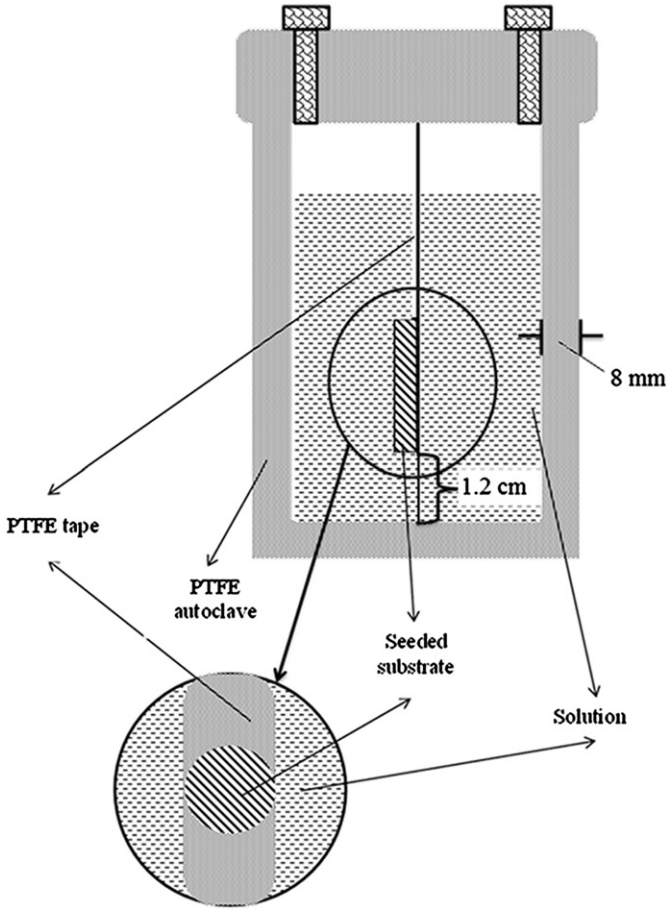

Fig. 1. Schematic diagram of the autoclave used for synthesis of membranes.

3. Characterization of seed and zeolite membranes

3.1. SEM and XRD

The crystal structures of the synthesized zeolite powder and also the as-synthesized zeolite membranes were characterized by X-ray diffraction (XRD). XRD measurements were performed by a Siemens powder diffractometer using Cu K_α radiation operating at 40 kV and 30 mA. The morphology and thickness of the as synthesized zeolite membranes were observed by scanning electron microscopy (SEM). The size and morphology of the synthesized A-type zeolite seeds were also determined by SEM. The SEM images were obtained using a Vega Tescan scanning electron microscope.

3.2. Pervaporation tests

Performance of the synthesized membranes for dehydration of ethylene glycol (EG) was characterized by pervaporation (PV) experiments. The experimental set up used for evaluation is schematically depicted in Fig. 2. In the PV experiments, the permeate side was evacuated and permeate vapor was condensed using a cold trap immersed in liquid nitrogen. The PV performance of the membranes was determined using separation factor (α) and permeation flux (J). Separation factor for component i over component j and total permeation flux (J) were respectively defined as:

$$\alpha_{i/j} = \frac{y_i/y_j}{x_i/x_j} \quad (1)$$

$$J = \frac{w}{A \times \Delta t} \quad (2)$$

where x_i and x_j are weight fractions of component i and component j in feed mixture; y_i and y_j are corresponding weight fractions in permeate; w is the total weight of permeate, kg; Δt is the experiment time, h; and A is the effective area of the membrane, m^2. Total permeate flux (J) was calculated using the weight of permeate collected over a specific period of time in the cold trap, and the effective membrane area. Composition of the permeate was determined using an accurate refractometer (DR-A1).

The PV experiments were conducted using a feed solution of 90:10 wt% EG/water mixture at 70 °C. To further investigate the quality of synthesized

membranes, PV experiments were carried out for dehydration of a feed solution of 90:10 wt% ethanol/water mixture at 70 °C.

4. Results and discussion

4.1. Sub-micro seeds of A-type zeolite

The XRD patterns of the synthesized powder are presented in Fig. 3. The main peaks associated with A-type zeolite can be observed in the XRD patterns.

To further evaluate the morphology and size of the synthesized zeolite seeds, SEM images were obtained. Fig. 4 shows the SEM micrographs of the synthesized Sub-micro seeds. As observed from the SEM images, the crystallite size is in the rang of 150 to 350 nm and the crystals are spherical.

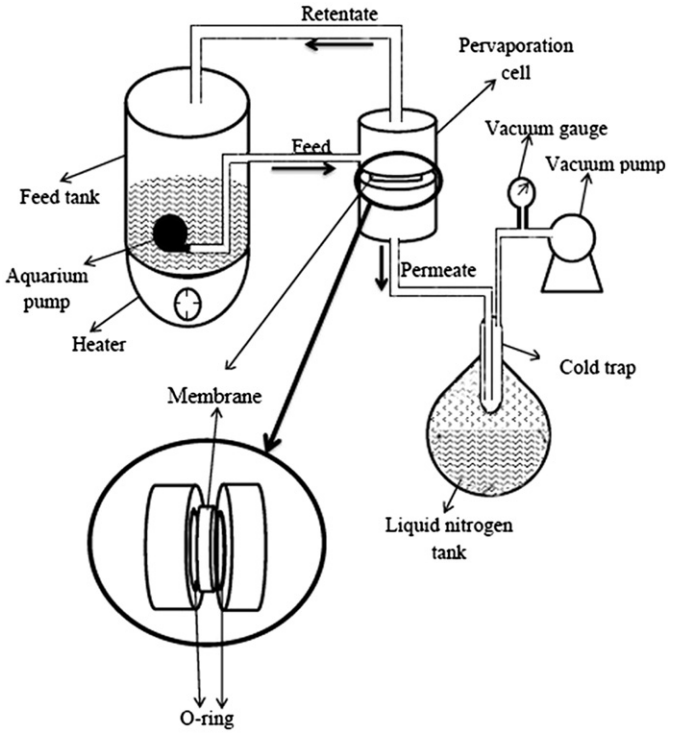

Fig. 2. Schematic diagram of the apparatus used for pervaporation experiments.

4.2. A-type zeolite membranes

4.2.1. Seeding

In order to synthesize a high quality A-type zeolite membrane, a thin and uniform layer of A-type zeolite on the support surface must be formed. To do this, some criteria must be taken into account. First, the seeds should be dispersed homogeneously on the support surface and the amount of seeds on the support surface should be optimal. Otherwise, the as-synthesized A-type zeolite membrane becomes uneven or too thick [37].

Fig. 5a shows SEM image of the support surface used in the synthesis of zeolite membranes. Fig. 5b represents SEM image of the seeded support. It confirms that the support is covered homogeneously with the seeds.

Another criterion for synthesis of high quality zeolite membranes is consistency between the seed size and the

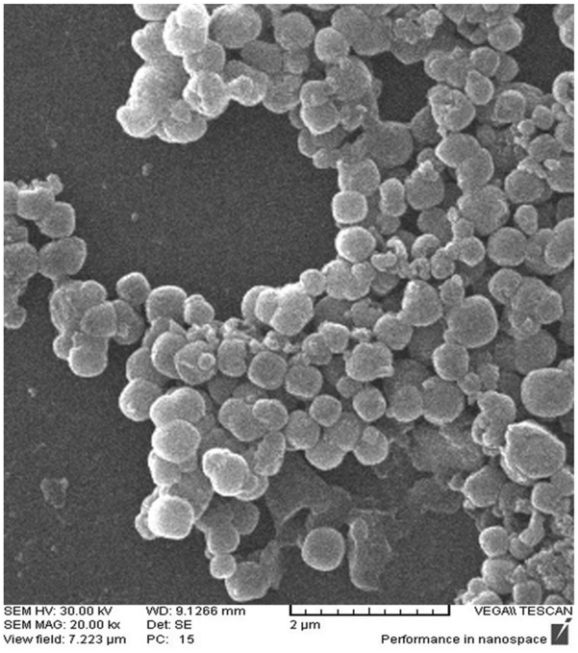

Fig. 4. SEM images of the synthesized A-type zeolite seeds.

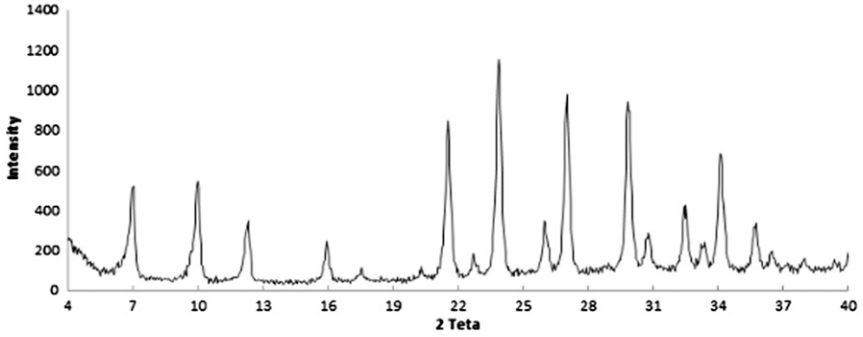

Fig. 3. XRD patterns of the A-type zeolite seeds.

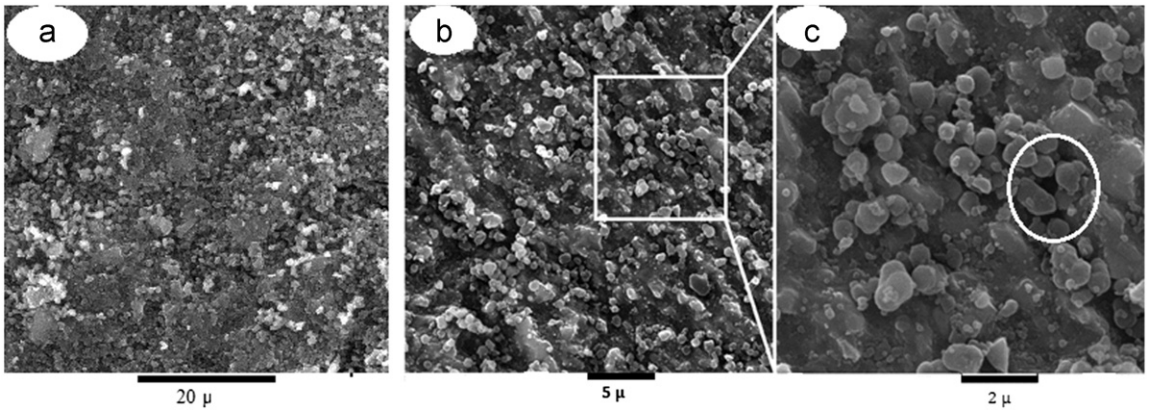

Fig. 5. SEM top view images of the seeded support.

Table 1
Synthesis conditions of A-type zeolite membranes and their PV performances.

No.	Synthesis time (h)	Synthesis temperature (°C)	Flux (kg m^{-2} h^{-1})	Separation factor
M1	1.15	80	6.6	–
M2	2	80	2.99	69
M3	3	80	3.42	637
M4	4	80	2.33	1,294
M5	1.15	90	7.10	–
M6	2	90	4.92	1,225
M7	3	90	3.71	10,000
M8	4	90	2.57	10,000
M9	1.15	100	6.10	–
M10	2	100	4.03	10,000
M11	3	100	3.53	10,000
M12	4	100	2.36	10,000
M13	1.15	110	6.00	–
M14	2	110	2.95	10,000
M15	3	110	2.00	10,000
M16	4	110	3.15	254

support pore size. To obtain high permeance, deep formation of A-type zeolite crystals inside the support pores should be avoided. Therefore, the seeds should not penetrate into the support pores and the seed size should be in consistence with the pore size of the support [26,38]. Fig. 5c shows SEM image of the seeded support. It shows that there is consistency between the seeds and the support so that the seeds do not penetrate into the support pores.

4.2.2. Effect of synthesis temperature on performance of A-type zeolite membranes

Synthesis temperature has significant effect on the rate of zeolite membrane formation. High temperature can accelerate the zeolite membrane synthesis. However, high temperature may change crystalline structure of zeolites in hydrothermal method [39]. Thin zeolite layer is favorable for zeolite membranes. Pinholes and cracks maybe presented in thin zeolite layers resulting in separation factor reduction. Therefore, optimum conditions must be chosen for thin zeolite layers and high quality zeolite membranes, i.e. without pinholes and non zeolitic pores.

Table 1 show membranes synthesized at different temperatures for different times. The performance of synthesized zeolite membranes including separation factor and permeation flux for dehydration of EG in PV is also listed in Table 1.

Generally, increasing synthesis temperature decreases permeation flux (see Table 1). As it can be observed, at synthesis temperature of 80 °C for different synthesis times, the membranes exhibit poor separation performances. The low selectivity at the synthesis temperature of 80 °C may be due to the incomplete formation of zeolite layer on the support surface. On the other hand, the low permeation flux at 80 °C may be attributed to the plugging of support pores by non-permeable phases [40].

Generally, increasing synthesis temperature increases separation factor (see Table 1). Fig. 6 shows SEM images of the membranes synthesized at different temperatures (80–110 °C) for 2 h. As it can be observed in Fig. 6 M2, at

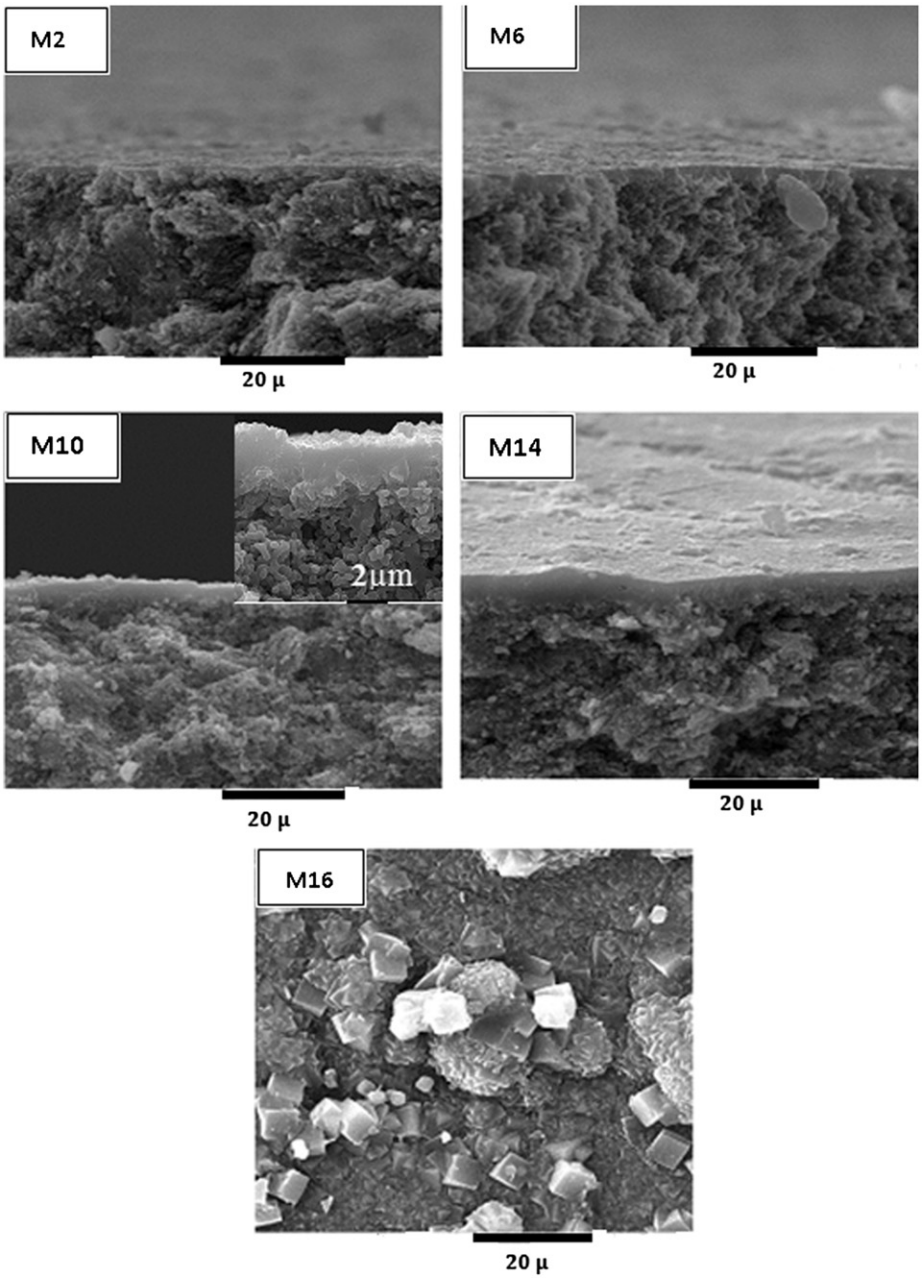

Fig. 6. Effect of synthesis temperature on secondary growth of A-type zeolite membranes.

synthesis temperature of 80 °C, the zeolite layer is not formed properly. Further increasing synthesis temperature to 90 °C results in formation of the relatively uniform zeolite layer, but some defects and pinholes exist in the zeolite layer (see Fig. 6, M6). The uniform and dense zeolite layer with high crystallinity can be formed when synthesis temperature increases further to 100 °C (see Fig. 6, M10 and Fig.7, M10). Much further increasing temperature to 110 °C causes the uniform and defect free zeolite layer is damaged (see Fig. 6, M14). The permeation of this membrane is lower than that of the membrane synthesized at 100 °C. This trend can be attributed to the membrane thickness.

Extending synthesis time to 4 h at synthesis temperature of 110 °C (M16) results in lower separation factor and higher permeation flux. This can be due to the formation of another zeolite phases such as NaX or P. Also, formation of pinholes and cracks at higher synthesis temperatures for longer synthesis times may be responsible for the poor performance (see Fig. 6, M16). The results confirm that the synthesis temperature of 100 °C is the optimum synthesis temperature for preparation of A-type zeolite membranes.

4.2.3. Effect of synthesis time on performance of A-type zeolite membranes

According to Table 1, increasing synthesis time leads to reduction of permeation flux except for M2 and M16 samples as discussed earlier. Fig. 8 presents SEM images of the zeolite NaA membranes synthesized on α-alumina

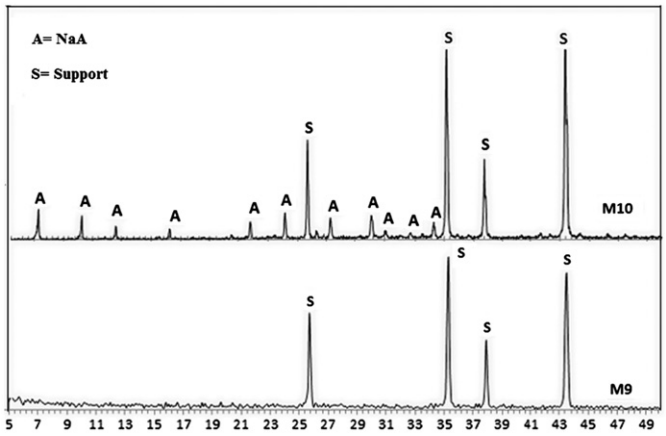

Fig. 7. XRD patterns for as-synthesized zeolite A-type membrane layers (M9 = 1.15 h, M10 = 2 h).

supports for different synthesis times. As observed, at synthesis temperature of 100 °C, for synthesis time of 1.15 h the membrane layer with thickness of 2 μm and low crystallinity is formed (see Fig. 8, M9-C and Fig. 7, M9). Surface SEM image of the membrane also confirms this observation (see Fig. 8, M9-S). By extending synthesis time to 2 h, the defect free membrane layer with thickness of 2.5 μm is formed (see Fig. 6, M10-C and Fig. 8, M10). The spherical crystals with crystal size between 0.5 and 1 μm can be observed for synthesis time of 2 h (see Fig. 8, M10-S). With further increasing synthesis time to 3 h, a zeolite layer with thickness of 8 μm is formed with the almost cubic crystals (see Fig. 8, M11-S and M11-C).

The results suggest that high quality A-type zeolite membranes can be hydrothermally synthesized at temperature of 100 °C for synthesis time of 2 h. This short synthesis time can

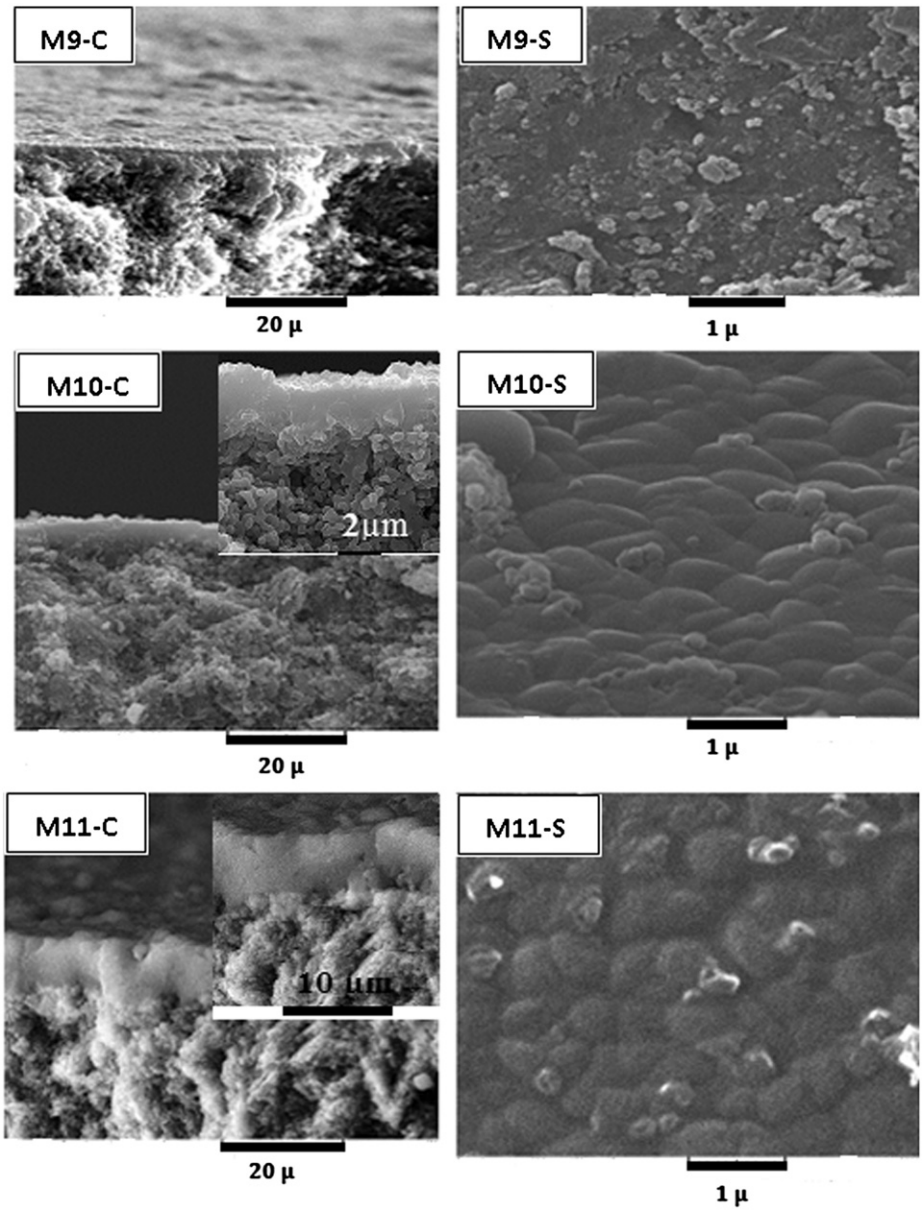

Fig. 8. Effect of synthesis time on secondary growth of NaA zeolite membrane (M9 = 1.15 h, M10 = 2 h, M11 = 3 h).

Table 2
Comparison of the PV properties of the as-synthesized NaA zeolite membranes prepared in this study with literature data.

Supports	T_S(°C)	t_s (h)	T_F (°C)	w_F, H_2O (wt%)	Flux (kg m^{-2} h^{-1})	αA/B	Ref.
α-Al_2O_3	100	24	70	25	0.11	10,000	[41]
α-Al_2O_3	85	6.5	50	8	1.2	8,500	[42]
α-Al_2O_3	100	3.5	75	10	3.45	2,330	[43]
Porous metal	100	—	75	10	4	10,000	[40]
CHFs	100	2	75	10	8	1,000	[44]
Mullite	105	4	70	10	2.6	10,000	[45]
α-Al_2O_3	100	4	75	10	5.6	5,000	[21]
α-Al_2O_3	100	4	75	10	8.5	10,000	[46]
α-Al_2O_3	100	2	70	10	11.14	10,000	This study
T_S=synthesis temperature		t_s=synthesis time		T_F=feed temperature		w_F H_2O=H_2O concentration in feed	

be attributed to the sub-micro powders used as seeds for synthesis of the membranes. Smaller seeds need shorter synthesis time compared with bigger seeds to form uniform and dense zeolite layer on the surface of support.

The reasons why small seeds are suitable for synthesis of high quality zeolite membranes can be summarized as follows: (i) more uniform seeding of support. For large seeds, the gravity field makes the seeding non-uniform. (ii) Better adhesion of seeds to support. During seeding process, when support is being pulled away from seed suspension, large seeds may be detached from support. (iii) Better coverage of seeds. Surface coverage of large seeds on support is much less than that of small seeds. (iv) Faster rate of zeolite membrane growth. For small seeds, large specific surface area provides adequate contact area with nutrient solution. Therefore, compared with large seeds, much more crystal nuclei surround small seeds leading to formation of dense zeolite layers [26].

4.3. Separation performance of synthesized membranes for ethanol dehydration

To further investigate the separation performance of synthesized A-type membranes, dehydration of ethanol was also carried out. Dehydration of ethanol is widely used to characterize zeolite membranes. PV dehydration of ethanol using the synthesized A-type zeolite membranes was carried out and the results were compared with the other works. Table 2 shows the results for dehydration of ethanol using A-type zeolite membranes synthesized by the hydrothermal method reported in literature and the obtained results. As shown, the membrane synthesized in this work exhibits high separation performance towards ethanol dehydration and total flux and separation factor of 11.14 kg m^{-2} h^{-1} and 10,000 are obtained, respectively. Separation factor and flux data obtained in this study are higher than other reported data.

5. Conclusions

Optimization of A-type zeolite powders and membranes were studied experimentally in this work. High quality membranes were prepared by adjusting the synthesis parameters including synthesis time and synthesis temperature. Sub-micro powder of A-type zeolite with high crystallinity and crystallite size ranging from 150 to 350 nm was synthesized at 60 °C for 11 h and characterized by solid characterizations including XRD and SEM. The crystal morphologies were spherical at these conditions. The sub-micro powders were then used as seeds for synthesis of zeolite membranes. The most efficient A-type zeolite membranes were obtained for synthesis time of 2 h at synthesis temperature of 100 °C. The PV performance of the membranes were evaluated via dehydration of EG. The synthesized membranes showed high permeation flux of 4.03 kg m^{-2} h^{-1} and high separation factor of more than 10,000 for a mixture of 10:90 (wt%) water/EG mixture at 70 °C. It was concluded that synthesis time of A-type zeolite membranes can be significantly reduced using sub-micro size A-type zeolite seeds. Since the performance of zeolite A-type membranes for dehydration of ethanol is an indicator of membrane quality, the synthesized zeolite membranes were characterized in PV dehydration of 10:90 (wt%) water/ethanol at 70 °C. The membranes showed total flux and separation factor of 11.14 kg m^{-2} h^{-1} and 10,000, respectively. The results for dehydration of ethanol obtained in this study were higher than other results reported in literature.

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