

# Effects of synthesis parameters on zeolite membrane formation and performance by microwave technique

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Recently, zeolite membranes on porous supports have been extensively studied in the ethanol–water separation process for further use for gasohol production. This work focuses on a NaA membrane synthesized on an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> support via microwave hydrothermal treatment. Synthesis temperature and time, type of substrate, seed amount and seeding time for the layer growth of the membrane are considered. The formation of as-synthesized membranes is discussed according to observations by SEM and XRD. In addition, a preliminary study of the performance of the synthesized NaA zeolite membrane was conducted using the pervaporation technique. It was found that, for the synthesized continuous NaA membranes prepared using a 0.5  $\mu$ m NaA crystal seed concentration of 3 g/l via vacuum seeding, the optimum conditions were 363 K synthesis temperature for 15–20 min via microwave heating. The flux and the separation factor obtained were 1.6 kg/m<sup>2</sup> h and 1760.5, respectively, for the substrate without an intermediate layer. Interestingly, the substrate *with* an intermediate layer showed better flux and separation factor at 1.7 kg/m<sup>2</sup> h and 6532.7, respectively.

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**KEYWORDS:** zeolite membrane; thin film; morphology and pervaporation; microwave technique

## INTRODUCTION

Since the beginning of the 1980s, many attempts have been made to develop zeolite membranes for separation and catalysis applications.<sup>1,2</sup> Specifically, the NaA zeolite membrane has the potential to sieve out molecules in a continuous process due to its hydrophilicity, making the electrostatic interaction between ionic sites and the water

molecule stronger.<sup>3</sup> As a result, it has been commercially applied to alcohol dehydration and solvent dewatering.<sup>4</sup>

In general, for zeolite membrane synthesis, the composition of the synthesis mixture, the temperature during synthesis and synthesis duration are the main parameters for determining which zeolite phase will be formed.<sup>3</sup> For efficient separation, the membrane must be free of any defects which could provide alternative transport pathways to the zeolite pores.<sup>4</sup> According to well-defined properties for separation purposes, controlling the thickness and texture of coatings is a significant task in determining the performance of the system. Thin and continuous zeolite films are required for some applications, especially those related to membrane separation. The film should be as thin as possible for optimal performance, and free of any pinholes. Nair and Tsapatsis<sup>3</sup> fabricated a zeolite NaA membrane using *in-situ* membrane growth or a secondary (or seeding) growth technique. They found that, for the latter technique, zeolite crystals were deposited on the support surface before exposure to hydrothermal growth conditions, whereupon the seed crystal

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grew into a continuous film. In the absence of a seed layer, a discontinuous NaA membrane is able to be formed on the support surface, as detected by a discontinuous layer.<sup>5,6</sup> Many researchers have reported the seed coating technique on the support substrate,<sup>7–11</sup> and one simple and effective method is vacuum seeding to coat a seeding layer onto the porous  $\alpha$  alumina support.<sup>12</sup>

Zeolite NaA can be synthesized by either microwave<sup>13</sup> or conventional hydrothermal<sup>14</sup> techniques. However, the microwave synthesis of zeolite has many advantages, viz. much shorter synthesis time, narrow zeolite particle size distribution and high purity.<sup>15</sup> In this study, the alumina substrate is firstly coated with a seed layer using the vacuum seeding method before microwave heat treatment. The effect of seeding parameters, viz. seed amount, seeding time and synthesis parameters, e.g. microwave temperature and time, on membrane perfection and thickness was investigated. The synthesized membrane was characterized using SEM and XRD. The type of substrate needed to improve the performance of the membrane by pervaporation was also considered.

## EXPERIMENTAL

### Materials

Sodium hydroxide (NaOH) from Lab-Scan Co. Ltd was used as the base catalyst. Fumed silicon dioxide ( $\text{SiO}_2$ , 474 m<sup>2</sup>/g surface area, 0.007  $\mu\text{m}$  average particle size) and aluminum hydroxide hydrate [ $\text{Al}(\text{OH})_3 \cdot x\text{H}_2\text{O}$ , 51 m<sup>2</sup>/g surface area] were purchased from Aldrich Co. and used as starting materials. Two types of substrates were used as supports, porous  $\alpha$  alumina tubes (11 mm outer diameter, 9 mm inner diameter, 6 mm length, 0.3  $\mu\text{m}$  pore radius on average with 38% porosity) and porous  $\alpha$  alumina tubes coated with an  $\alpha$ -alumina intermediate (0.06  $\mu\text{m}$  pore size) on the top layer. The intermediate layer was formed by dip-coating the porous  $\alpha$  alumina into a submicron-sized of alumina suspension for 1 and 5 s, denoted by C1 and C2, respectively. All supports were supplied by the National (Thailand) Metal and Materials Technology Center (MTEC).

### Equipment

All samples were characterized using scanning electron micrographs (SEM) on a Jeol 5200-2AE (MP15152001) and wide angle X-ray diffractograms (WXR) on a D/MAX 2000 series Rigaku X-ray diffractometer using  $\text{CuK}\alpha$  as the source. Membrane synthesis was performed using microwave on a Milestone, Ethos SEL Spec 1000 W and 2450 MHz. The separation factor from the quantity of water and ethanol was determined using gas chromatography (Win Lab III, Perichorm). Surface area and pore volume of samples were determined by Physisorption of nitrogen at temperature of 77 K using an Autosorb I instrument.

**Table 1.** JCPDS file for Zeolite A (dehydrated)

$2\theta$	$d$ (Å)	$I$	$2\theta$	$d$ (Å)	$I$
7.20	12.278	100	35.83	2.506	0.7
10.19	8.682	50	36.60	2.456	2.8
12.49	7.088	23.2	38.09	2.363	1.4
16.14	5.491	19.7	40.23	2.242	0.5
17.70	5.012	1.9	41.61	2.170	1.4
20.46	4.341	3.0	42.29	2.137	1.4
21.41	4.151	0.8	42.96	2.106	1.6
21.72	4.093	9.6	43.61	2.075	1.0
24.04	3.702	13.7	44.27	2.046	2.2
26.17	3.405	0.6	47.42	1.917	1.8
27.18	3.281	9.1	48.03	1.894	1.9
30.01	2.978	11.1	52.73	1.736	4.3
30.90	2.894	1.8	54.41	1.686	1.3
32.62	2.745	1.5	56.10	1.626	1.0
33.45	2.679	1.0	57.67	1.598	1.6
34.26	2.618	8.2	58.74	1.572	1.0

### NaA zeolite membrane synthesis

The porous  $\text{Al}_2\text{O}_3$  tube was cleaned and washed with deionized water by ultrasonication for 15 min to remove any dirt from the surface. The cleaned support was dried at 363 K for 24 h and calcined at 673 K for 3 h to burn off any impurities on the support surface before coating with seed crystals. Membrane synthesis with various conditions, as listed in Table 2, was carried out by first coating the NaA zeolite crystal on the alumina substrate, followed by immersing the coated substrate into a vessel containing the solution mixture for further microwave heat treatment.

Vacuum seeding<sup>12</sup> was used to coat the NaA seed onto the support tube. The suspension was prepared by dispersing 2–4 g NaA zeolite ( $\sim 0.5 \mu\text{m}$  particle size) synthesized using the  $\text{SiO}_2 : \text{Al}_2\text{O}_3 : 3\text{Na}_2\text{O} : 410\text{H}_2\text{O}$  formula, as described in Kuanchertchoo *et al.*,<sup>16</sup> in 1000 ml of water with ultrasonic treatment. The seeding layer was coated onto the outer surface of the support tube using 0.0325 MPa for 1, 2 and 3 min. The seeded support was dried in air at 383 K for 24 h before characterization using SEM.

The seed-coated  $\text{Al}_2\text{O}_3$  tubes were placed vertically in a Teflon vessel containing the solution ( $5\text{SiO}_2 : \text{Al}_2\text{O}_3 : 50\text{Na}_2\text{O} : 1000\text{H}_2\text{O}$ ), prepared according to Xu *et al.*,<sup>17</sup> in the microwave chamber. After crystallizing at 363 K for a desired time, repeated synthesis (multistage synthesis) was carried out to improve the quality of the NaA zeolite membrane. The synthesized membrane was washed several times with deionized water and was then dried in air at 363 K for a few days before characterizing using SEM, XRD and BET. The JCPDS card of zeolite A (dehydrated) is displayed in Table 1.

### Pervaporation

The efficiency of the synthesized membrane on the support tube was determined from the separation factor and the water

flux by the pervaporation, using an ethanol–water mixture (95:5) at a feed rate of 0.898 l/min, 10 mmHg vacuum pressure and 343 K. The quantities of water and ethanol were determined using gas chromatography. The GC column used was an HP plot Q capillary column at a TCD detector temperature of 250 °C and oven temperature 160 °C and with a helium gas flow rate of 180 kPa.

## RESULTS AND DISCUSSION

As mentioned previously, membrane performance, viz. flux and separation factor, is membrane-formation dependent. High flux and good separation were obtained from using a thin and continuous zeolite film. Thus, the following study of the effects on the membrane formation clearly reflect the membrane's performance.

### Effect of seed amount

The most important factor in zeolite membrane preparation is the fabrication of a very thin film with a defect-free membrane layer in order to obtain a high efficiency of flux and separation. Thus, the preparation of a small and uniform zeolite particle is crucial. Generally, to get a good seeding layer, the seed must be as small as possible, but big enough to avoid penetrating the support pore, which would lead to a poor separation factor and low flux.<sup>12</sup> Huang *et al.*<sup>12</sup> found that the presence of NaA crystal seeds on the support surface could promote zeolite formation. The secondary growth technique, based on the pre-deposition of a dense seed layer, appears to be a possible method for preparing a defect-free membrane with suitable separation properties.<sup>3</sup>

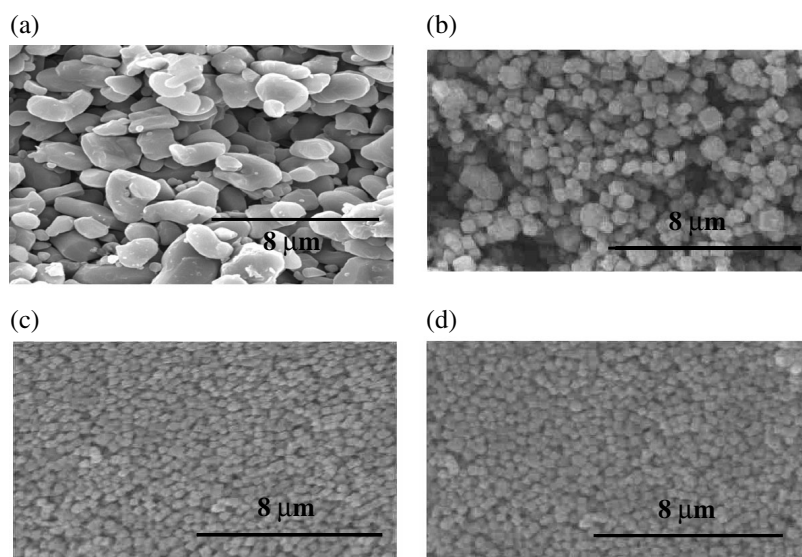
The seed amount has a significant effect on the quality of the seeding layer and the performance of the zeolite membrane.

Seed amounts of zeolite, containing 2, 3 or 4 g/l of seed in water were studied at a coating pressure of 0.0325 MPa for 1, 2 or 3 min coating time. With 1 min seeding time, the support surface was not completely covered (not shown), whereas with 2 or 3 min coating time there was enough time for homogeneous coating on the substrate. The results from the SEM (Fig. 1) show that NaA crystal seeds were continuously coated on the substrate surface when 3 or 4 g/l of the seed amount was used.

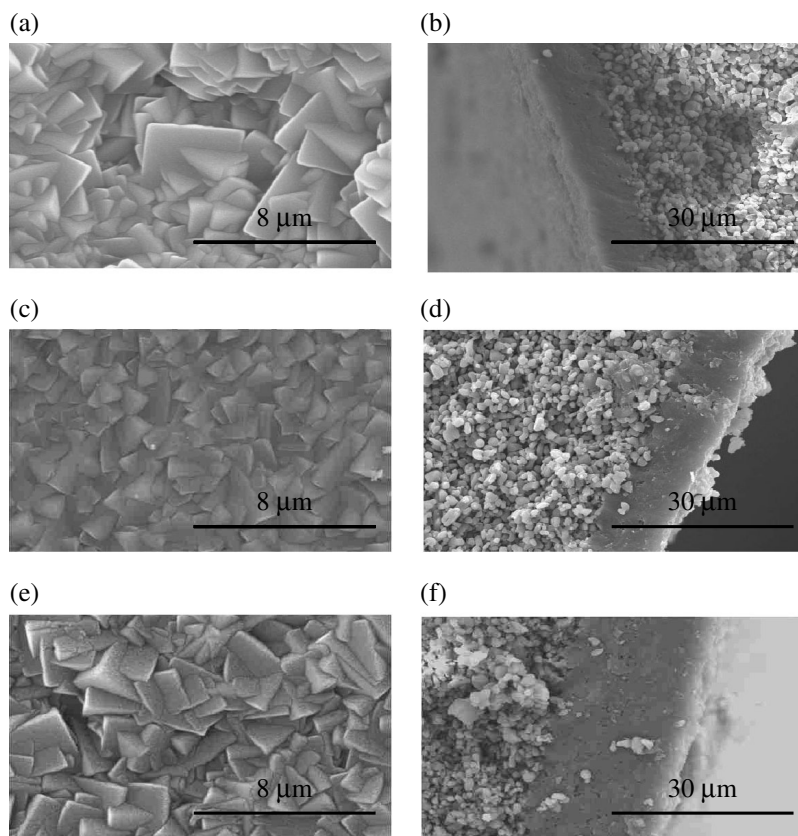
Figure 2(a–f) shows the SEM images of the NaA zeolite membrane synthesized by microwave at 363 K for 20 min and using various seed concentrations of 2, 3 and 4 g/l. The separating flux and factor are summarized in Table 2. It can be seen from the results that, with 2 g/l, the substrate was discontinuously covered and the crystals were not well inter-grown. In the cross-section view [Fig. 2(b)] of the membrane, defects and holes are also observed, which cause poor membrane performance. The separation factor and the water flux were 673.6 and 2.4 kg/m<sup>2</sup> h, respectively (see Table 2). When the seed concentration was increased to 3 g/l, a purer and denser NaA zeolite membrane was formed, providing higher performance (separation factor = 1288.0). The membrane thickness in the cross-section view is about 10 µm. Up to 4 g/l seed concentration, the NaA zeolite membrane was too thick (~20 µm) and had some cracks, resulting in poor performance and consequently giving a separation factor and a water flux of 677.0 and 1.4 kg/m<sup>2</sup> h, respectively.

### Effect of seeding time

Huang *et al.*<sup>12</sup> reported that seeding time has the same effect as seed concentration on the properties of the seeding layer and the zeolite membrane. In our case as well, the seeding



**Figure 1.** SEM micrographs of (a) AE substrate and (b)–(d) NaA crystal seed on substrate using 2, 3 and 4 g/l seed concentrations, respectively, for 3 min.



**Figure 2.** SEM micrographs of NaA zeolite membrane synthesized on substrate using seed concentrations of (a, b) 2, (c, d) 3 and (e, f) 4 g/l, vacuum seeding for 3 min under 363 K microwave radiation for 20 min.

time influenced the seeding layer, the thickness of which was increased with seeding time. At a longer seeding time, the seeding layer becomes too thick and has cracks, causing poor performance of the synthesized membrane. On the other hand, if the seeding time is too short (for example, 1 min), the seeding layer will not be continuous, as confirmed by the SEM results (not shown), and the performance of the NaA membrane formed (separation factor = 1125.6, flux = 1.2 kg/m<sup>2</sup> h) is not as good as those seeded for 2 or 3 min. The 3 min seeding time was, in fact, too long, giving a thick membrane and probably causing some cracks (separation factor = 1338.5, flux = 1.4 kg/m<sup>2</sup> h). A coating time of 2 min seems to be the best choice for obtaining a better quality seeding layer. Moreover, the separation factor of the NaA zeolite membrane, in this case, was improved. The membrane thickness was about 9.2 μm, determined from a cross-sectional view. The flux was as high as 1.6 kg/m<sup>2</sup> h and the separation factor was 1790.5. When the seeding time was too long (4 min), the seeding layer was too thick, resulting in too thick membrane (12.3 μm). Some cracks were observed (not shown). The separation factor and the water flux were 76 and 2.38 kg/m<sup>2</sup> h, respectively.

The pore volumes and surface areas of synthesized, continuous NaA membranes prepared using a 0.5 μm NaA

crystal seed concentration of 3 g/l via vacuum seeding for 1, 2 and 3 min at 363 K synthesis temperature for 15 min microwave heating time are listed in Table 3.

Interestingly, the surface area of NaA zeolite seed (0.5 μm) obtained was only 6.9 m<sup>2</sup>/g. This result was also observed by Suzuki *et al.*<sup>18,19</sup> They indicated that the abnormal phenomena may have been caused by a considerably reduced accessibility of adsorbate, nitrogen, to the internal structure of NaA zeolite at liquid nitrogen temperature<sup>19</sup> because molecules with a kinetic diameter larger than 0.36 nm are not adsorbed.<sup>20</sup>

The surface area and the pore volume of ground NaA zeolite membrane on an alumina tube prepared using 2 min seeding time (giving a separation factor of 1790) was increased by about 18.8 and 20%, respectively. The surface areas of those prepared using 1 and 3 min seeding time (giving separation factors of 1125.5 and 1338.5, respectively) were also increased by 6.3 and 12.5%, respectively. However, the pore volumes were reduced by 13 and 15%, respectively. It can be concluded that the separation factor of NaA zeolite membrane increased with the pore volume capacity.

### Effect of microwave time

Generally, NaA zeolite can be synthesized by different routes, namely, autoclave, electrophoresis and microwave

**Table 2.** Pervaporation results at various conditions

Conditions	Substrate wall thickness (mm)	Zeolite layer ( $\mu\text{m}$ )	Flux ( $\text{kg}/\text{m}^2 \text{ h}$ )	Separation factor
<i>Seed concentration</i> (3 min seeding time at 363 K microwave temperature for 20 min)				
2 g/l	1.5	9.5	2.4	673.6
3 g/l	1.5	10.5	2.05	1288.0
4 g/l	1.5	20.5	1.4	677.0
<i>Seeding time</i> (3 g/l seed concentration at 363 K microwave temperature for 15 min)				
1 min	1.5	7.9	1.2	1125.6
2 min	1.5	9.2	1.6	1790.5
3 min	1.5	9.6	1.4	1338.5
<i>Microwave synthesis time</i> (at 3 min seeding time and 363 K microwave temperature)				
3 g/l seed concentration				
20 min	1.5	10.5	2.1	1288.0
25 min	1.5	13.8	0.9	169.5
4 g/l seed concentration				
20 min	1.5	20.5	1.4	677.0
10 min	1.5	16.6	1.2	1125.6
<i>Microwave synthesis temperature</i> (2 g/l seed concentration for 3 min and 15 min microwave synthesis time)				
333 K	1.5	5.5	3.1	5.1
348 K	1.5	6.5	2.9	7.6
363 K	1.5	9.2	1.6	1760.5
378 K	1.5	8.5	17.9	2.1
<i>Substrate type</i>				
with C1 intermediate layer	1.5	10.7	0.8	4203.2
with C2 intermediate layer	1.0	7.4	1.7	6532.7

**Table 3.** Nitrogen physisorption of NaA zeolite,  $\text{Al}_2\text{O}_3$  substrate and NaA zeolite membrane which were prepared by using 3 g/l seed concentration with 1, 2 and 3 min seeding time and heating at microwave temperature 363 K for 15 min

Samples	Surface area ( $\text{m}^2/\text{g}$ )	$V_p$ ( $\text{cm}^3/\text{g}$ )
NaA zeolite (0.5 $\mu\text{m}$ )	6.9	$1.9 \times 10^{-2}$
$\text{Al}_2\text{O}_3$ substrate (pore diameter 0.3 $\mu\text{m}$ )	1.6	$4.5 \times 10^{-3}$
NaA zeolite membrane with 1 min seeding	1.7	$5.1 \times 10^{-3}$
NaA zeolite membrane with 2 min seeding	1.9	$5.3 \times 10^{-3}$
NaA zeolite membrane with 3 min seeding	1.8	$5.2 \times 10^{-3}$

$V_p$  = total pore volume for pores with diameter less than 1694.2 Å at  $P/P_0 = 0.98854$ .

techniques. Comparing with autoclave, a dense membrane occurs at longer synthesis time<sup>12,21,22</sup> because nuclei are not formed on the support simultaneously due to a low dissolution of gel on the surface and low heating rate. Thus, the zeolite crystals formed are not uniform in size and the membrane obtained will be thick.<sup>23</sup> In this study,

NaA zeolite membrane was synthesized using microwave technique due to the advantages of using much shorter synthesis time, giving homogeneous heat delivered directly to materials through molecules with electromagnetic field. Continuous NaA zeolite membranes were synthesized by secondary growth on an external surface of  $\alpha$ -alumina tubular supports, using the seeding technique to direct the membrane synthesis towards a desired phase, and a 3 g/l seed concentration. The synthesized membrane was prepared at a 363 K microwave heating temperature for 15–20 min synthesis time, because a synthesis time longer than 20 min, for example 25 min, generated a film that was too thick and had some cracking, resulting in poor separation performance. The support surface was completely covered with NaA zeolite crystals at the synthesis times of 15 and 20 min with a membrane thickness of 9.6 and 10.5  $\mu\text{m}$ , respectively.

NaA zeolite membranes synthesized using a seed concentration of 4 g/l for 3 min seeding time showed that after a 10 min microwave heating treatment, the substrate was continuously covered with a NaA zeolite crystal layer with a thickness of 16.6  $\mu\text{m}$ . Using the same conditions, the surface of the seed-coated support was completely covered with a NaA zeolite membrane layer with a thickness of 20.5  $\mu\text{m}$  after heating for 20 min. However, the membrane did contain some cracks.

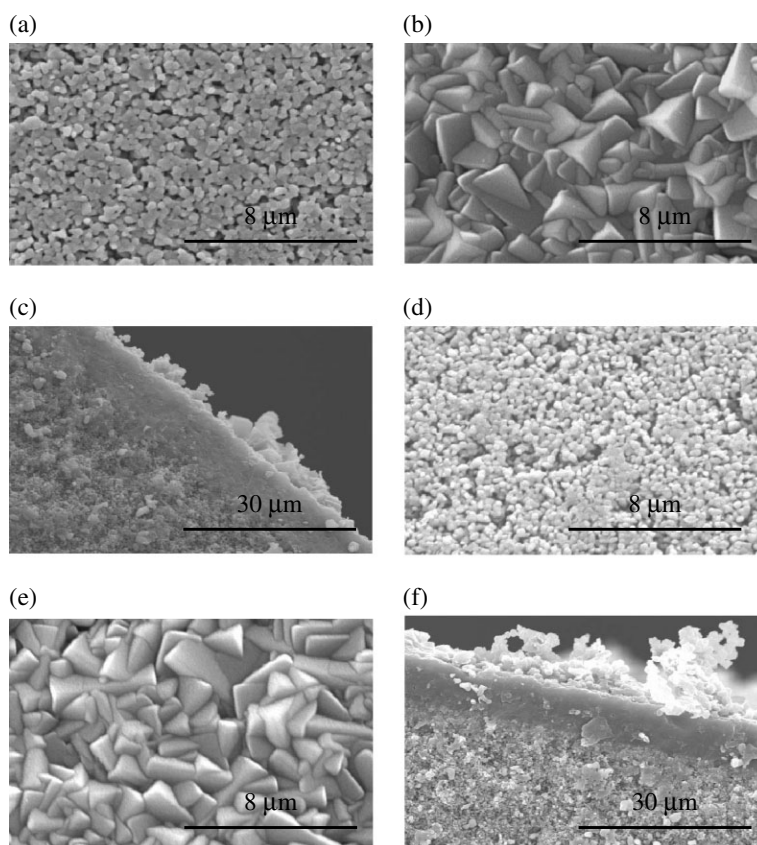
### Effect of microwave temperature

The hydrothermal treatment conditions used for the preparation of the zeolite membrane affect not only the primary microstructure parameters, such as membrane thickness and crystal size, but also the zeolite morphology, crystal intergrowth and apparent surface porosity.<sup>21</sup> In this work, the effect of synthesis temperature (at 333, 348, 363 and 378 K) was studied by fixing the formula of the membrane precursor solution ( $5\text{SiO}_2 : \text{Al}_2\text{O}_3 : 50\text{Na}_2\text{O} : 1000\text{H}_2\text{O}$ ), the amount of the NaA seed crystals (2 g/1000 ml of water), the synthesis time (15 min) and the seeding time (3 min). The SEM results (not shown) revealed that, at 333 K, the substrate was covered with a discontinuous NaA zeolite crystal seed layer having a NaA crystal size of between 0.5 and  $1.5\text{ }\mu\text{m}$  and some holes within the NaA crystal layer. Moreover, the side view of the membrane revealed that the layer was not well inter-grown, indicating the poor quality of the synthesized membrane, as can also be confirmed by the results obtained from the separation factor and the flux (Table 1). A microwave temperature of 348 K provided a better membrane surface, with a 6–7  $\mu\text{m}$  thickness. Defects and holes, however, were still detected, giving a low separation factor of 7.6. After heating at 363 K,

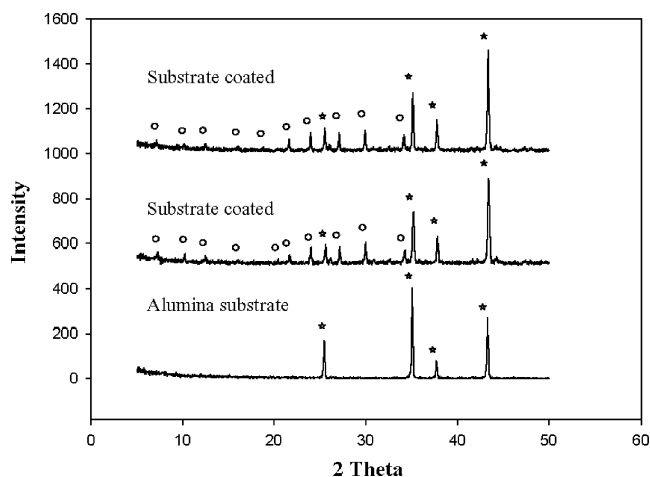
the substrate was completely covered with well-inter-grown NaA crystals and a continuous NaA zeolite layer ( $9.2\text{ }\mu\text{m}$ ). The flux and separation factor were improved to  $1.6\text{ kg/m}^2\text{ h}$  and 1760.5, respectively. When the temperature was increased to 378 K, the NaA crystals on the substrate were dissolved and transformed into hydroxyl-sodalite zeolite crystals. The dissolution of NaA zeolite on the support surface increased the concentrations of mono- and dimeric-silicate ions near the surface, and reached the concentration required for the formation of other types of zeolites, as discussed by Wong and co-workers.<sup>23</sup> This phenomenon expectedly generates the poor separation performance.

### Effect of type of substrates

Many researchers<sup>24–27</sup> are interested in the preparation of a thin mesoporous layer (or intermediate layer) on macroporous substrates to improve the performance of zeolite NaA by preventing the penetration of organic components into the substrate.<sup>22</sup> In this research, the intermediate layer is thus applied to possibly obtain a higher performance of the NaA zeolite membrane. SEM images of the intermediate top layers of substrates C1 and C2 are shown in Fig. 3(a and d), respectively. The synthesis was performed at 363 K



**Figure 3.** SEM micrographs of (a) C1 intermediate layer; (b, c) surface and cross-sectional view of NaA zeolite membrane synthesized on C1 intermediate layer; (d) C2 intermediate layer; and (e, f) surface and cross-section views of Na A zeolite membrane synthesized on C2 intermediate layer.



**Figure 4.** XRD patterns of NaA zeolite coating on C1–C2 intermediate layer substrates; (\*) alumina substrate; (o) NaA Zeolite. (a) Alumina substrate, (b) NaA zeolite on substrate C1 and (c) NaA zeolite on substrate C2.

for 20 min because, when using a shorter time ( $<20$  min), a high separation factor and flux could not be obtained (not shown).

Figure 3(b, c, e–f) shows SEM images of the membrane synthesized on the intermediate layer of a macroporous substrate under microwave radiation at 363 K for 20 min. The substrates were completely covered with NaA zeolite having a thickness of 10.7 and 7.35  $\mu\text{m}$  for C1 and C2, respectively. Moreover, only NaA zeolite was formed on the support, as confirmed by XRD (Fig. 4), showing the phase of the NaA zeolite membrane synthesized on different intermediates, compared with the microporous substrate and synthesized LTA.<sup>16</sup> The dense layer of the NaA zeolite was observed by SEM, taken from a cross-sectional view; see Fig. 3(c and f).

It is clear that the smooth surface of the intermediate layer provided a greater advantage for NaA zeolite membrane formation. The performance of the synthesized membrane was indeed improved, as clearly seen in Table 2. The separation factors of the membrane synthesized on the intermediate layers C1 and C2 were 4203.2 and 6532.7, respectively. The reason for this improvement was probably due to the smaller pore size of the intermediate layer, making the zeolite seeds unable to fill in the pores. As a result, the seeds were homogeneously attracted to the surface over the whole area, forming a much smoother and more uniform seed layer. It was found that the wall thickness of the C2 tube before coating with the intermediate layer was thinner than that of the C1 tube. This might be why the higher flux of the membrane synthesized on the intermediate C2 substrate was observed; that is, optimization of the support characteristics (for example a thinner layer, high porosity and large pores) affects the opportunities for the achievement of higher fluxes.<sup>28</sup>

## CONCLUSIONS

NaA zeolite membranes were successfully prepared by vacuum seeding and microwave heating techniques under different conditions on alumina substrates with and without an intermediate layer. Synthesis parameters, such as seed amount, seeding time and synthesis time, affected the film thickness after synthesis and the separation performance. The higher the film thickness, the lower the flux obtained. The higher temperature also caused an impurity phase on membrane during synthesizing, resulting in low separation performance. The optimum conditions were found to be 3 g/l of seed solution containing 0.5  $\mu\text{m}$  zeolite crystal seed at 0.0325 MPa seeding pressure for 2 min seeding time and 363 K microwave temperature, for 15 min on the supports without an intermediate layer and 20 min on the one with an intermediate layer. The flux and the separation factor obtained were 1.6  $\text{kg}/\text{m}^2 \text{ h}$  and 1760.5, respectively, for the substrate without an intermediate layer. The substrate *with* an intermediate layer showed a better flux and separation factor, 1.7  $\text{kg}/\text{m}^2 \text{ h}$  and 6532.7, respectively, owing to the smoother surface resulting from the more homogeneous seeding on the substrate and the thinner membrane. The intermediate layer of C2 with the thinner wall thickness (1.0  $\mu\text{m}$ ) gave higher flux (1.69  $\text{m}^2/\text{g}$ ) than that of C1 (0.83  $\text{kg}/\text{m}^2 \text{ h}$ ).

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