# CHAPTER III
# EXPERIMENTAL

## 3.1 Materials

Fumed silica ($SiO_2$ surface area 474 $m^2/g$, average particle size 0.007 μm) and aluminum hydroxide ($Al(OH)_3.x\ H_2O$, surface area 51 $m^2/g$) were purchased from Sigma Chemical Co. Ltd., and used as received. Sodium hydroxide (NaOH) and nitric acid ($HNO_3$) from Labscan Co. Ltd., were used as received. Porous α alumina tubes ($\alpha$-$Al_2O_3$, 11 mm outer diameter, 9 mm inner diameter, 0.3 μm pore radius in average with 38% porosity) were used as supported and supplied by National Metal and Materials Technology Center (MTEC). NaA zeolite (home-made NaA zeolite, particle size < 1 μm) was used as crystal seed.

## 3.2 Equipment

Scanning electron micrographs (SEM) were obtained on a JEOL Model JSM 5410 using a voltage of 15-20 KV at 1500 to 7500 magnification. The samples were stuck to brass stubs and coated with gold by vapor deposition. Wide angle x-ray diffractograms (WXRD) on D/MAX 2000 series of Rigaku X-ray diffractometer system were analyzed to identify the zeolite NaA pattern using Cu Kα as a source performed in a range of 5-50 degree/min and a 0.02 degree scan step. Divergence and scatter slits at 1 degree together with 0.3 mm receiving slit were set. Microwave hydrothermal treatment was performed on MSP 1000, CEM Corporation Spec 1000 W and 2450 MHz. A sample was heated in a Teflon vessel using organic digestion mode and time to temperature program. Vacuum seeding was also used to coat NaA seed on a support tube using 0.0325 MPa for designed time.

## 3.3 Methodology

### 3.3.1 Preparation of NaA Zeolite for Using as the Crystal Seed

The mixture for NaA zeolite synthesis was prepared using the $SiO_2$: $Al_2O_3$ : $3Na_2O$ : $410H_2O$ (Kuanchertchoo *et al.*, 2006) formula.

a. The solution was firstly prepared by dissolving 0.39g of sodium hydroxide in 12 ml deionized water
b. 0.0975 g of fumed silica was added into this solution with stirring.
c. When the solution was homogeneous, 0.2536 g of aluminium hydroxide was then added to the mixture solution, followed by vigorously stirring to produce a homogeneous solution.
d. The solution is finally added with 3 wt% NaA seed crystal. This solution was vigorously stirred overnight (aging) before microwave heating at 60°C for 10 h. The synthesized NaA zeolite was washed with water until pH is neutral and dried in oven at 90°C for 24 h.

### 3.3.2 Preparation of α $Al_2O_3$ Tube (Zeolite Membrane Supported Material)

a. The surface of the support tube was polished with a 600 grit-sand paper.
b. The support was then cleaned and washed with deionized water by ultrasonication for 15 min to remove loosen particles created during polishing.
c. Cleaned support was dried at 90°C for 24 h and calcined in air at 400°C for 3 h with heating and cooling rates of 4°C/min and 10°C/min, respectively, to burn off impurity on support surface before coating with seed crystals.

### 3.3.3 Preparation of the Seeded Support

a. The colloidal suspension was firstly prepared by dispersing 1-3 g NaA zeolite seed (from home-made NaA zeolite, particle size < 1 μm) in 1000 mL of deionized water with ultrasonic treatment for 15 min.

b. After 24 h of settlement, the top part of the solution containing amorphous components was removed. The crystal at the bottom part was dried in oven.

c. The dried NaA zeolite seed (1-3 g.) was dispersed in 1000 mL of deionized water with ultrasonic treatment for 15 min. After 3 h of settlement, the top clear part was removed to use as seed colloidal suspension.

d. The seeding layer was coated on the outer surface of support tube by vacuum seeding using 0.0325 MPa for designed time, followed by drying in air at 90°C for 3h.

3.3.4 Preparation of the Solution for A-type zeolite membrane synthesis

This solution is prepared using the $5SiO_2$:$Al_2O_3$:$50Na_2O$:$1000H_2O$ (Xu *et al.*, 2001) formula.

a. The solution was firstly prepared by dissolving 44.44 g of sodium hydroxide in 200 ml deionized water

b. 1.7 g of aluminum hydroxide was added into this solution with stirring.

c. When the solution was clear, fumed silica is then added to the mixture solution with vigorously stirring to produce a clear and homogeneous solution again.

3.3.5 Zeolite Membrane Synthesis

The seeded support was placed centrally and vertically in a Teflon vessel to avoid any precipitation of zeolite crystals onto the support during the membrane synthesis. The synthesis solution was carefully poured into the vessel without touching the support. The vessel was then sealed. After crystallizing by microwave treatment at a designed temperature for a designed time, the synthesized membrane was washed several times with deionized water until the pH of the washing is neutral, followed by drying in air at 90°C for 24 h.

Conventional technique is similar to microwave technique. Teflon vessel in a stainless steel autoclave was used to replace the microwave chamber at 60°C synthesis temperature. After crystallization at 60°C for the designed time, the

as-synthesized sample was washed and dried similar to those obtained from microwave technique.

Electrophoresis technique is also similar to the autoclave technique, except that it is applied by 2V voltage. After crystallization at 60°C for the designed time, the as-synthesized sample was washed and dried.

3.3.6 Characterization

a. The structure of the synthesized membrane is determined using XRD.
b. The morphology and the thickness of the synthesized membrane are examined using SEM.