

CHAPTER III EXPERIMENTAL

3.1 Materials

All adsorbent materials were provided by UOP LLC. NaX-zeolite, AgX-zeolite and silicalite were used as adsorptive fillers incorporated into cellulose acetate (acetyl content of 39.5%) which purchased from East Chemical Company . Acetone used as solvent in cellulose acetate was obtained by Labscan and Fisher Scientific. Gases used in this study were ethane, ethylene, propane, propylene and nitrogen. Ethane (purity>99.5%) and ethylene (purity>99.99%) were provided by National Petrochemical Co.Ltd. Commercial grade of propane (purity > 99.95%) and propylene (purity > 99.5%) from Metheson were provided by UOP LLC. Nitrogen (purity> 99.999%) was obtained from Prax Air.

All adsorbent materials-NaX-zeolite, AgX-zeolite and silicalite were studied the selectivity of hexene to hexane to select adsorbent materials for making mixed matrix membrane. They were tested equilibrium adsorption process by gas chromatography (GC) and were calculated the equilibrium selectivity as mathematical derivations shown in the Appendix.

3.2 Membrane Preparation

There were three types of mixed matrix membranes, which were studied in this work. They were NaX-zeolite/cellulose acetate, AgX-zeolite/cellulose acetate and silicalite/cellulose acetate mixed matrix membranes. These membranes were prepared by solution casting method. All types were prepared at 5, 10 and 20 wt% of additive fillers by combination of 0.238, 0.50 and 1.125 grams of additive fillers for each respective weight in

acetone of 39.5 grams (AR grade, 99.6 wt%) and stirred about 3 hours. This was done to ensure removal of air in the pores of adsorbent materials. Subsequently, 4.5 grams of cellulose acetate (acetyl content of 39.5%) was added to the additive filler-acetone suspension and stirred solution about a day to obtain homogeneous solution. The application of a partial vacuum ensured the removal of air bubbles entrained during cellulose acetate incorporation.

The resulting suspension was coated on the top horizontal surface of a clean glass plate in the size of 8*10 inches by using a casting knife. The casted film was cut in circular having a diameter of 7.5 cm and the thickness was about 60-70 μm . The use of a casting knife allows for the formation of a uniform, even film of an appropriate polymer solution on the glass plate. The casting knife consists of a steel blade, resting on two runners, arranged to form a precise gap between the blade and the plate on which the film is cast. The thickness is set by the use of two micrometers. Once the film has been cast, a portion of the acetone was allowed to evaporate slowly until a film was formed on the upper surface of the solution. After an additional set time of 20-45 seconds, the membrane was submerged into an ice water bath (4-5 $^{\circ}\text{C}$) about 3 to 4 minutes. The film was then soaked in a hot water bath at 65-75 $^{\circ}\text{C}$ for one hour to remove residual acetone. The membrane was dried between paper towels and two glass plates to prevent curling of the membrane. Figure 3.1 shows the preparation procedure of NaX/cellulose acetate, AgX/cellulose acetate and silicalite/cellulose acetate mixed matrix membrane. The pure cellulose acetate membranes were prepared in a similar manner without the addition of adsorptive filler.

Scanning electron microscopy (SEM) was used at UOP to analyze the presence of the incorporated zeolites in the cellulose acetate matrix. Bulk structures were observed cross sectional view for membrane fractured in liquid nitrogen. Then, this membrane was prepared by attaching it to the specimen stub and applying a conductive surface coating with gold (Sawyer and Grubb,

1996). Only AgX-zeolite/cellulose acetate mixed matrix membrane was selected to analyze membrane morphology. In cases of NaX-zeolite/cellulose acetate and silicalite/cellulose acetate mixed matrix membranes, materials of NaX-zeolite and silicalite are similar to AgX-zeolite therefore the morphologies of these adsorptive fillers in the same polymer matrix should be similar.

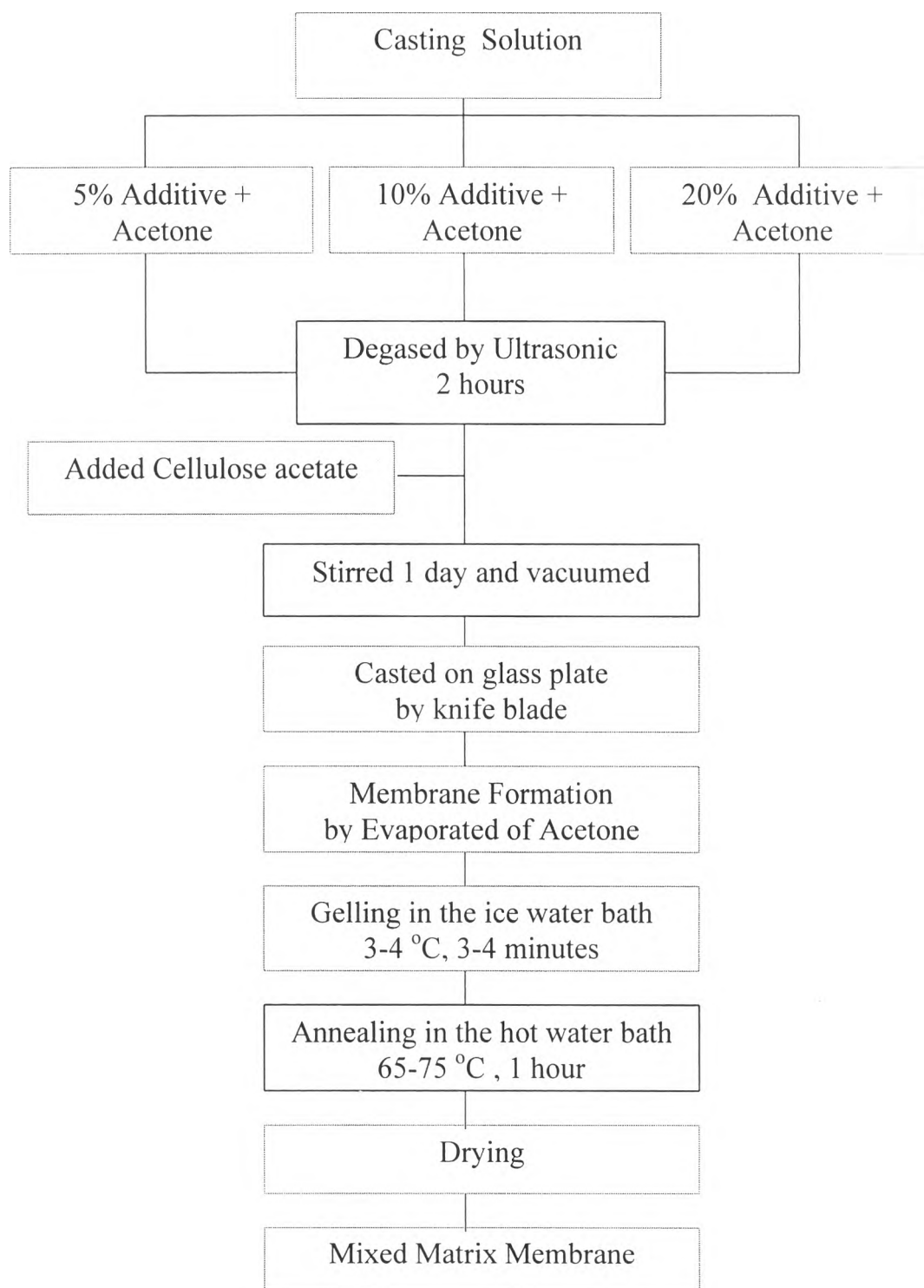


Figure 3.1 Mixed matrix membrane preparation procedure

3.3 Design and Experimental Setup

The overall set-up for the experiments is shown in Figure 3.2. Gas cylinders containing the various test gases (propane, propylene, and nitrogen) were connected to the test cell. The prepared mixed matrix membrane, having a diameter of 7.5 cm, was placed in the test cell with an O-ring forming a seal around the edge as shown in Figure 3.3. A porous metal plate supported the membrane. The flowrate of gas was measured with a soap-bubble meter.

There were 7 steps to test a membrane:

- 1) A pressure test with N_2 was performed to check for leaking of gas in the experimental setup.
- 2) The pressure of N_2 was increased to 100 psig and the flux was measured to check for pinhole defects.
- 3) The system was allowed to come to steady state over approximately 10 hours, and the steady state flux was measured.
- 4) Shut off the N_2 and degas the setup at the permeate side for about 5 minutes. Subsequently, the C_3H_8 was connected and the system was purged for 5 minutes by opening the outlet valve at the retentate side.
- 5) The outlet valve was closed at the retentate side and C_3H_8 was allowed to permeate through membrane about 10 hours to displace residual N_2 gas in the membrane phase and to achieve steady state flux.
- 6) Disconnect the C_3H_8 and degas the C_3H_8 (same as step 4) by vacuum pump at the permeate side about 5 minutes, follow by purging with N_2 for 5 min.
- 7) Finally, the outlet valve was closed and N_2 flux was measured as in step 5. Steps 6 and 7 were repeated for C_3H_6 and N_2 , respectively. For each membrane, it took 4 days to finish a test.

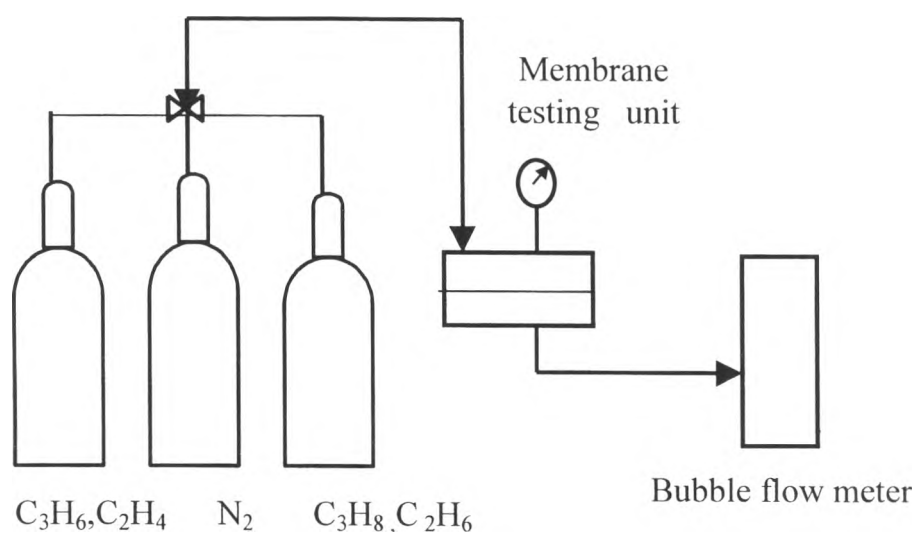


Figure 3.2 Schematic diagram of the experimental setup

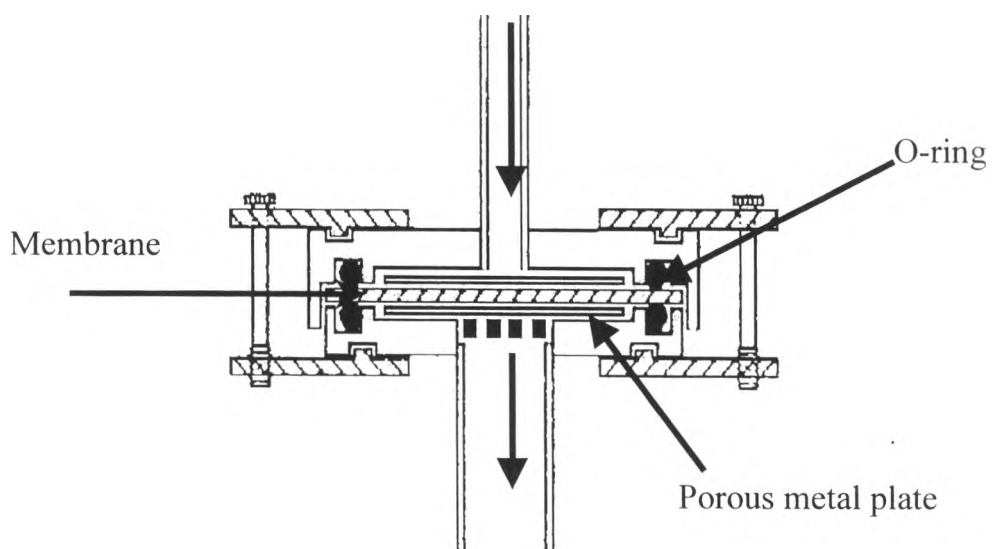


Figure 3.3 The cross section of the membrane testing unit