

CHAPTER IV

EXPERIMENTS

4.1 Chemicals

1. Calcined Alumina A12 : Alcoa, USA.
2. Calcined Alumina A16SG : Alcoa, USA.
3. Aluminium sec-butoxide : Aldrich Chemical Company, Inc., USA.
4. Copper oxide : Fluka Chemika, Switzerland.
5. Titanium oxide : Fluka Chemika, Switzerland.
6. Magnesium oxide : Fluka Chemika, Switzerland.
7. Alginic acid : Sigma Chemical Company, Inc., USA.
8. Polyvinyl alcohol : Aldrich Chemical Company, Inc., USA.
9. Dispex A40 : Loxley (Bangkok) Ltd.
10. Hydrochloric acid : Ajax Chemicals, Australia.
11. Silicone antifoam : Osi Specialties (Thailand) Ltd.
12. Nitric acid : Ajax Chemicals, Australia.
13. Sodium dihydrogen phosphate : Ajax Chemicals, Australia.
14. Disodium hydrogen phosphate : Ajax Chemicals, Australia.
15. Soluble starch : E.Merck, Germany.
16. 3,5-dinitrosalicylic : Sigma Chemical Company, Inc., USA.

17. Sodium hydroxide : J.T.Baker, Inc., USA.
18. Sodium potassium tartrate : Ajax Chemicals, Australia.
19. Boric acid : E.Merck, Germany.
20. Iodine : Ajax Chemicals, Australia.
21. Potassium iodide : Fluka Chemika, Switzerland.

4.2 Equipments

1. Stirrer : Heizung, Germany.
2. Ball mill
3. Ultrasonic bath : Transsonic 460, Germany.
4. Cabolite : Nabertherm LHT model 16/R17, USA.
5. Sol-gel equipment
6. Filtration unit
7. X-ray Diffractrometer : PW 1730/10, Holland.
8. Universal Testing : Lloyd 2000R, USA.
9. Specific Surface Area Analyzer : Pore Sizer 9320, USA.
10. Specific Surface Area Analyzer : ASAP 2000, USA.
11. Scanning Electron Microscope : Jeol JSM-5410 LV, Japan.

4.3 Experimental Procedures

This study was divided to four sections: consist of preparation supported and intermediated layer as Sittinun's Method [2], preparation membranes by using sol-gel method, characterization membranes, and separation α -amylase enzyme by using these membranes. These are explained in the following.

4.3.1 Supported and Intermediated Layer Preparation

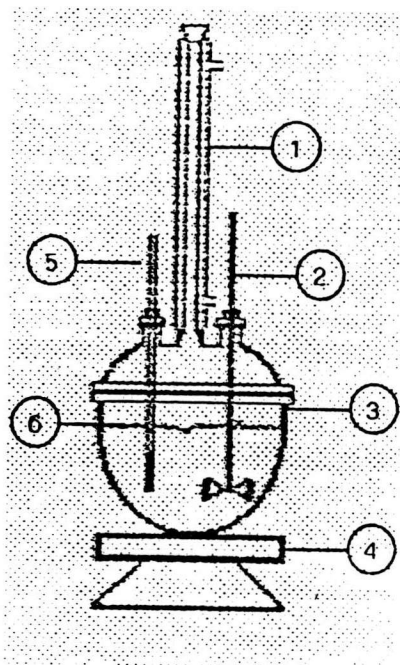
In this experiment, the slip of supported layer was prepared from Alumina A12, sodium alginate, sintering flux (ratio of CuO : TiO₂ : MgO is 1:1:0.05 by weight), dispex, and water in percent by weight of 55.56, 0.0556, 0.3817, 0.2222, and 43.78, respectively. This slip was milled for 3 hours, and casted it in mold for 8 minutes. Then, the obtainable tube was sintered at 1300 °C for 2 hours.

The slurry of intermediated layer which consists of Alumina A16SG, Polyvinyl alcohol, and water in percent by weight of 28.67, 0.0287, 71.30, respectively, 1 M Hydrochloric acid 5 ml, and anti-foam, was casted inside of the supported tube for 30 seconds. These coated tubes were sintered at 1200 °C for 30 minutes.

4.3.2 Sol-Gel Layer Preparation

Figure 4.1 shows the equipment of sol-gel. To make sol solution, 1 mole of Aluminium sec-butoxide (ASB) was put into 100 mole of water at 80-85 °C with vigorous stirring. The reaction was allowed to continue for 2 hours, then add 0.03 mole of nitric acid per mole of alkoxide into the reactants. Heating and stirring for another hour, after that the reaction vessel was opened to remove the butanol. A condenser was then positioned above of the vessel and the solution was refluxed for 16 hours. All procedures in this section were repeated by varying nitric acid (0.05, 0.07, 0.09, and 0.11 mole of acid per mole of alkoxide). The products were transferred to glass bottles and stored in the dark until needed.

After that, the tubes from section 4.3.1 were coated by those of sol solution on inner part of tubes. The coating time was varied from 10, 20, and 30 seconds. Then, sol-gel tubes were calcined at various temperatures and times (300 to 800 °C and 5 to 240 minutes, respectively).



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|----------------|-----------------|
| 1. Condenser | 2. Stirrer |
| 3. Sol Vessel | 4. Hot Plate |
| 5. Thermometer | 6. Sol Solution |

Figure 4.1 Sol-Gel Equipment

4.3.3 Supported and Membrane Layer Characterization

4.3.3.1 Bulk Density, Apparent Porosity, and Water Absorption Measurement

Density, porosity, and absorption of support were measured by standard test method according to ASTM C20 (see calculation in Appendix A).

4.3.3.2 Strength Measurement

Modulus of rupture of support was measured by T-point bending method, using Lloyd 2000R (see calculation in Appendix A).

4.3.3.3 Shrinkage Measurement

Shrinkage of support was observed by measurement technique, using the measurement before and after sintering (see calculation in Appendix A).

4.3.3.4 Specific Surface Area, and Pore Size Distribution Measurement

Surface area and pore size distribution of the support layers and membranes were measured by mercury intrusion method, with mercury as the adsorbate using a micromeritics model Pore Sizer 9320 and BET method, with nitrogen as the adsorbate using a micromeritics model ASAP 2000 at liquid-nitrogen temperature, consecutively.

4.3.3.5 Morphology

The shapes and the distribution of the pores were observed by Scanning Electron Microscope (SEM): JEOL JSM-5410 LV.

4.3.3.6 *X-ray Diffraction Patterns*

The transition phase of the membranes were performed by X-ray Diffractometer (XRD): PW 1730/10.

4.3.4 *Rejection Performance of Membranes*

The prepared membranes which shown in Figure 4.2 were investigated their injection by using 0.2% wt. Polyvinyl alcohol (MW 70,000-100,000) under pressure and recirculation velocity of 0.3 bar and 1.66 m/sec, respectively for 120 minutes of batch filtration. Each made tubes has diameter of 0.6 cm. and 25.4 cm. of length which is equal to 47.88 cm² of filtration area. Then 7 tubes in one module, which shown in Figure 4.3, were used to separate PVA solution. (Figure 4.4, and 4.5 show apparatus of filtration unit and schematic diagram of filtration apparatus). 20 ml of permeate was taken for amount of PVA analysis (see the method of analysis in Appendix B). After that, these first layer membranes were coated again under the same conditions to obtain bi-layer membranes. The rejection performance of bi-layer membranes was also determined in the same fashion.

4.3.5 *α -Amylase Enzyme Separation*

α -amylase enzymes (BAN 240 L of Novo Nordisk) solution were concentrated under enzyme concentration, recirculation velocity, and pressure of

2.4 g/litre, 1.66 m/sec, and 0.3 bar, respectively. The permeates were taken for permeation flux and saccharifying amylase determination (see the determination and saccharifying amylase in Appendix C). This separation were repeated at various pressure, recirculation velocity, and enzyme concentration (0.4 and 0.5 bars, 1.04 and 1.43 m/sec, 1.2 and 1.8 g/litre, respectively).

4.3.5.1 cleaning and Regenerating the Filter

The porous membranes, that are composed of γ -alumina, can be cleaned by chemical reagent using acid and alkali. In this experiment, the used filters were washed with distilled water, dip them into chemical solution (0.5 to 2.0 % NaOH) for 12 to 24 hours before washed with distilled water again. The regenerated filter was confirmed for next usage by measuring permeation flux of distilled water which would be in the same range of the new one.

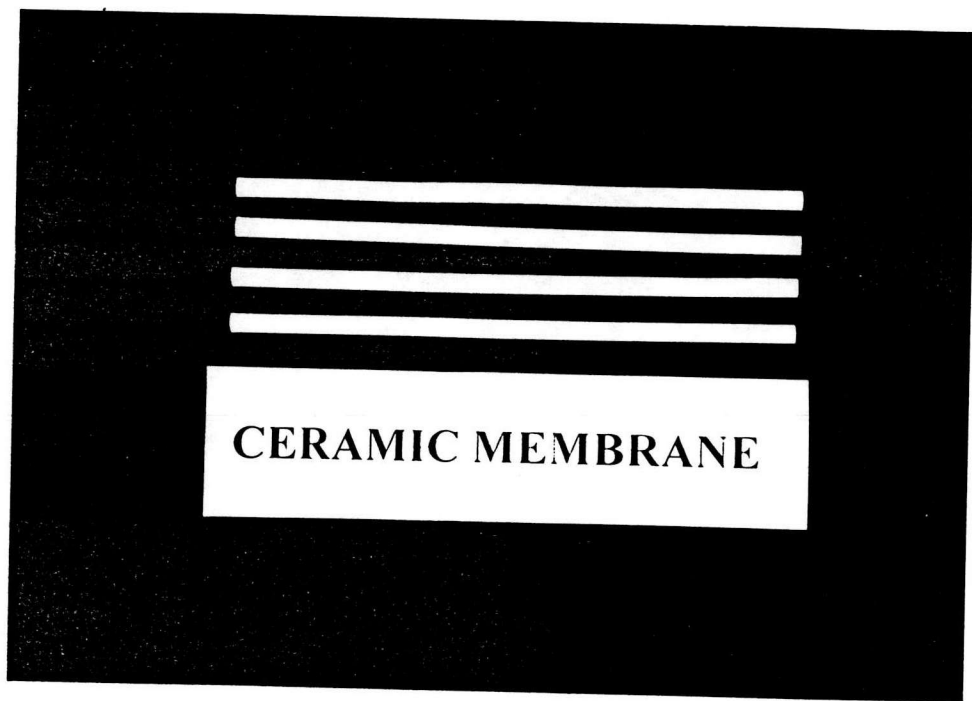
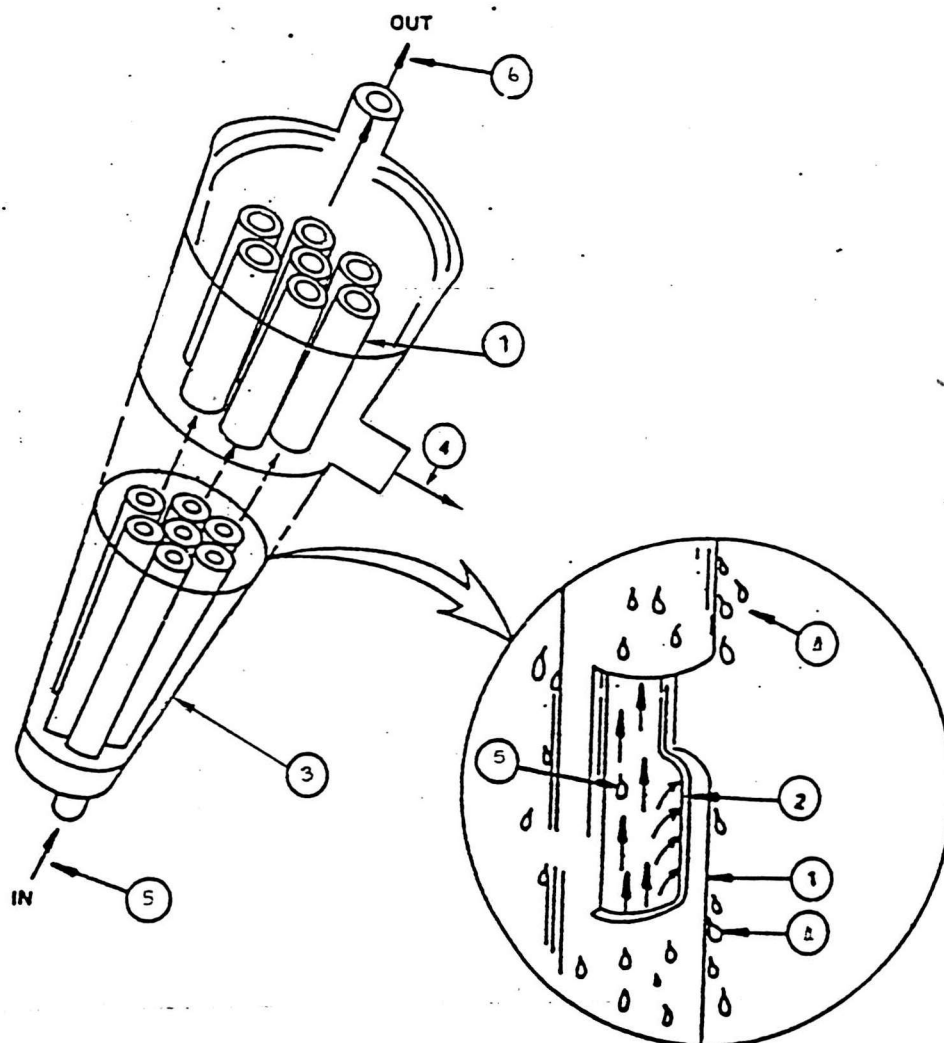


Figure 4.2 Photograph of Ceramic Filter



1. Supported Tube

2. Filtration Membrane

3. Module

4. Permeate

5. Feed Solution

6. Retentate

Figure 4.3 Schematic diagram of Ceramic Module

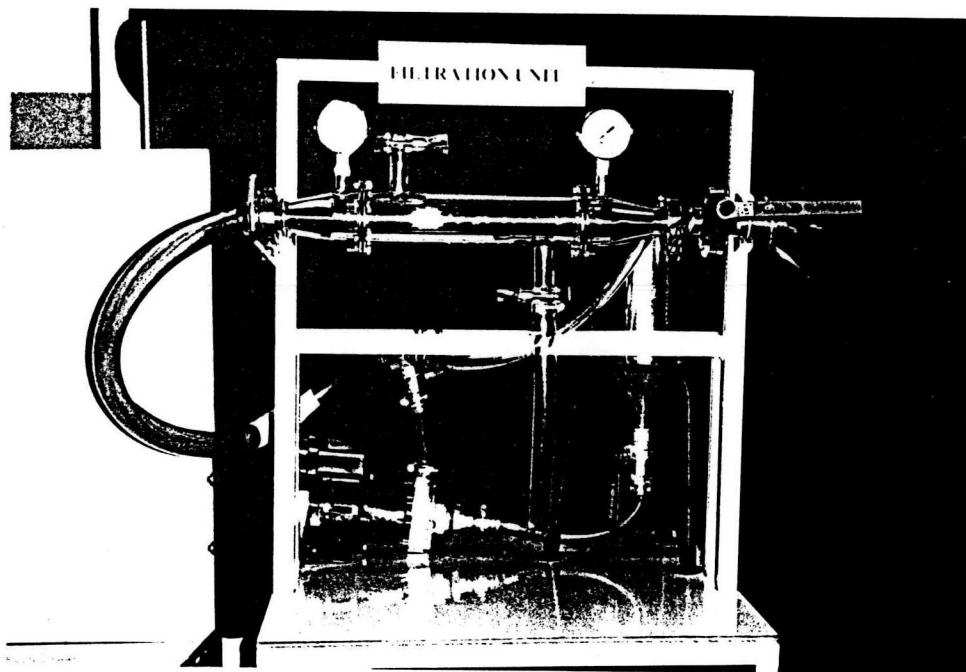
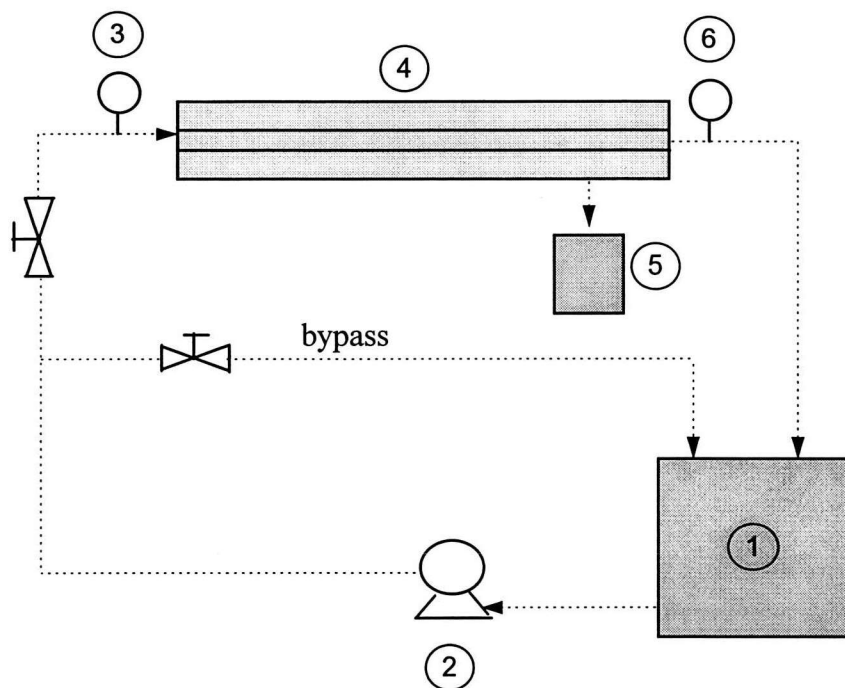


Figure 4.4 Experimental Apparatus of Filtration Unit



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|-------------------------|--------------------------|
| 1. Storage Tank | 2. Feed Pump |
| 3. Inlet Pressure Gauge | 4. Ceramic Module |
| 5. Permeate Tank | 6. Outlet Pressure Gauge |

Figure 4.5 Schematic diagram of Filtration Apparatus