CARBON DIOXIDE REMOVAL FROM NATURAL GAS BY USING SILICA MEMBRANE

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CHEMICAL ENGINEERING
UNIVERSITI TEKNOLOGI PETRONAS
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Nurul Safiah binti Mat Dagang

Dissertation submitted in partial fulfilment of
the requirements for the
Bachelor of Engineering (Hons)
(Chemical Engineering)

DECEMBER 2010

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CERTIFICATION OF APPROVAL

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Approved by,

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(A. P. DR M AZMI B BUSTAM @ KHALIL)

UNIVERSITI TEKNOLOGI PETRONAS
TRONOH, PERAK
December 2010

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

NURUL SAFIAH BINTI MAT DAGANG

ABSTRACT

The community is already paying high attention to the development of technologies especially in the natural gas processing. One of the attentions is on the development of membrane technology in the natural gas separation in order to get high quality of natural gas. Presence of carbon dioxide contributes to bad effect especially on the corrosion of equipments, pipelines and this can affect the overall operations. The focus on this technology is because of several advantages of membranes, such as efficient, simplicity, high selectivity and permeability, low cost requirement, stability under operating condition and easy to control leads to the attention of its development.

This project is to synthesize a silica membrane with high performance and high carbon dioxide/methane (CO_2/CH_4) selectivity and also to study the effect of pressure, inlet flow rate and dip coating duration on the separation performance. The silica membrane was prepared by the deposition of silica sol onto porous alumina support. The experiment started with the preparation of alumina support and percentage of porosity was calculated. The highest percentage porosity calculated was 21.89 % by using 20 drops (1.05 gram) of starch as a binder. Then, alumina support was dipped in the silica solution. In dip coating method, the times for dipping were varied for 1.25, 2.25 and 3.25 hours and permeability test was conducted. From the calculation, flux increases when the inlet flow rate and inlet pressure were increased. The highest flux was 1.911 cm^3[STP]/cm^2.s at 3.5 bar for 50.00 cm^3/s inlet flow rate. However, flux decreases as the dip coating hour increased (also when the silica thickness increases) and flux for carbon dioxide (CO_2) was higher compared to methane (CH_4).

The highest permeability was for carbon dioxide (CO_2) with 1.009660 cm^3[STP].cm/cm^2.bar.s at inlet pressure 1.5 bar for 50.00 cm^3/s inlet flow rate at 3.25 hours dip coating duration. The separation factor was the highest at 1.5 bar for 50.00 cm^3/s inlet flow rate at 3.25 hours dip coating with 1.4212 CO_2/CH_4 separation factor.

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CHAPTER 1

INTRODUCTION

1.1 Background

As natural gas becomes one of the high demand energy sources, the companies realize that natural gas needs to be commercialized in a high quality. All companies look forward for the technologies for the separation of contaminants in the natural gas. Natural gas is composed of main components which are methane, ethane, propane, butane and other hydrocarbon. The other components that contained in natural gas are carbon dioxide, hydrogen sulphide, nitrogen and water vapor which commonly known as contaminants.

Nowadays, the most demanding gas treatment facility is to remove carbon dioxide from the natural gas stream. Carbon dioxide is used by the food industry, the oil industry, and the chemical industry such as fire extinguisher, carbonated drinks and pest's control.[1] In the oil and gas industry, carbon dioxide is also used in gas injection as a commercial process for enhanced oil recovery (EOR).[2] However, this gas can give bad effect to the pipeline especially for the offshore facilities where the concentration of carbon dioxide is high in the well before separation process is done. Carbon dioxide needs to be removed to eliminate or reduce the bad consequences in daily operations.

Some natural gas contains a high percentage of carbon dioxide which is usually falls in range of 65%-70% and some are as high as about 80%.[3] Natural gas requires only below 2-3% of carbon dioxide according to pipeline specification.[4] Carbon dioxide falls into category of acidic acid. If there is water vapor in natural gas, carbon dioxide can react with the water vapor which will form carbonic acid. This acid is highly corrosive which can cause bad damage to the pipelines and equipments. This will lead to increase of maintenance and operating cost for the plant.

Many technologies have been developed to enhance the process of removing carbon dioxide from the natural gas stream. It includes the absorption processes which uses hot potassium carbonate solutions and Amine Guard-FSTM process (formulated solvents), cryogenic processes, adsorption processes, such as pressure swing adsorption (PSA), thermal swing adsorption (TSA) and iron sponge.[5] The latest technology that attracts many researchers to study is membrane technology. Membrane technology provides many advantages which are high efficiency, simplicity, high selectivity and permeability, compactness, stable under operating conditions, flexibility, and lower cost.[6] Therefore, it is important to find the best technology to ensure the quality of natural gas as well as the equipment, pipelines and overall operations.

In this project, silica membrane is prepared by dipping the alumina support disc from alumina oxide powder into the tetraorthosilicate solution. The dipping time is varied for 1.25 hours, 2.25 hours and 3.25 hours. The permeability test using carbon dioxide and methane gas is conducted to study the effect of inlet flow rate, inlet pressure and the dip coating duration on the separation performance. The permeability and CO_2/CH_4 separation factor are analyzed to study the separation performance.

1.2 Problem Statement

1) Low porosity percentage membrane from previous thesis which was 22.53 % by using starch.[7]
2) Low separation factor from previous thesis which was 1.37 by taking the ratio of permeability of CO_2 over CH_4.[7] The ideal separation factor is 1.66.[8] The previous thesis used duration of 1, 2 and 3 hours dipping duration.

1.3 Objectives and Scope of Study

The objectives of this study are:

a) To synthesize membrane with higher porosity than 22.53 % (up to 25-30 %).
b) To synthesize membrane that provides high permeability and selectivity higher than 1.37 (up to 1.66 ideal separation factor).
c) To evaluate the performance of the synthesized membrane.

The scopes of study are:

a) To research and study the suitable method to remove carbon dioxide from CO_2/CH_4.
b) To conduct experiment in the operating pressure 1 atm and operating temperature at room temperature, 25^oC.
c) To study the effect of inlet pressure, inlet flow rate and dip coating duration on the separation performance.

CHAPTER 2

LITERATURE REVIEW

2.1 Separation Process

Separation process is used to transform a mixture of substances into two or more distinct products. All separated products can be differed in terms of chemical or physical properties such as size or crystal modification. Separations process has three fundamental transport processes which are: [9]

a) Momentum transfer which occurs in moving media such as sedimentation and fluid flow.

b) Heat transfer which occurs in transferring of heat from one point to other point such as drying and evaporation.

c) Mass transfer which occurs in whether gas, solid or liquid phases such as absorption, membrane separation and adsorption.

Separation process can be classified into:

a) Evaporation: Evaporation of volatile solvent from a nonvolatile solvent, such as water and salt.

b) Drying: Removal of volatile liquid such as water from solid materials.

c) Distillation: Separates components of liquid mixture by boiling due to their vapor pressure differences.

d) Absorption: Removal of component from gas stream by treatment with liquid.

e) Membrane separation: Separation of a solute from a fluid by diffusion of this solute from a liquid or gas through semi permeable membrane barrier to another fluid.

f) Liquid-liquid extraction: Solute in solution is removed through contacting with another liquid solvent that is relatively immiscible with the solution.

g) Adsorption: Component is removed and adsorbed from a gas or liquid stream by a solid component.

h) Ion exchange: Ion-exchange solid is used to remove certain ions in solution.

i) Liquid-solid leaching: Treat a finely divided solid with a liquid that dissolves out and removes a solute contained in the solid.

j) Crystallization: Precipitate the solute from solution that is to be removed such a salt.

k) Mechanical-physical separations: Solids, liquids or gases are separated by mechanical means. Such as settling and filtration.

In this project, the main focus is on the membrane separation.

2.2 Membrane

Membrane is defined as a thin layer of material which separates two phases and acts as a semi permeable barrier. The separation occurs by controlling the rate of movement of various molecules between two liquid phases, two gas phases, or a liquid and a gas phase.[9] Membrane separations are classified by pore size and by the separation driving force. These classifications are gas diffusion in porous solid, liquid permeation or dialysis, gas permeation in membrane, reverse osmosis, ultra filtration membrane process, microfiltration membrane process and gel permeation chromatography.

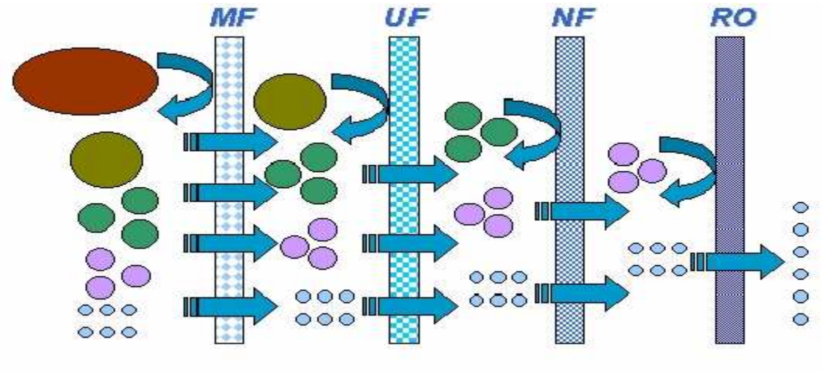


Figure 2.1: Type of membrane process. **Source: www.yale.edu*

2.3 Type of Membrane

Membranes can be divided into two types which are biological and synthetic membranes. A biological membrane or biomembrane is an enclosing or separating amphipathic layer that acts as a barrier within or around a cell such as the pleura that surrounds the lungs. It is almost invariably a lipid bilayer, composed of a double layer of lipid molecules which is usually phospholipids and proteins that may constitute close to 50% of membrane content. The size, charge, and other chemical properties of the atoms and molecules to cross it will determine whether they succeed in crossing the layer or not. Types of biological membranes are cell membranes and mucous membrane.[10]

Synthetic membrane is known as artificial membrane that is commonly used in reverse osmosis, microfiltration, ultrafiltration, pervaporation, dialysis, emulsion liquid membranes, membrane-based solvent extraction, membrane reactors, gas permeation, and supported liquid membranes.[11] Synthetic membrane is divided into two types which are **organic/polymer** (polyamide, cellulose acetate, polycarbonate) and **inorganic** (ceramic, glass, metallic, zeolites).

2.4 Application of Membrane

Membrane technology has seen a significant growth and increase in application for separation process. Several applications of membrane technology are as follow:

a) Reverse osmosis (RO) technology called nanofiltration (NF) which is also known as “membrane softening,” It has also been widely used for treatment of hard, high color, and high organic content feed water.[12]

b) Microfiltration (MF) and ultrafiltration (UF) membrane filtration technologies have emerged as viable options for addressing the current and future drinking water regulations related to the treatment of surface water and ground water.[12]

c) Membranes are also used in gas separation process. As mentioned before, in natural gas processing, applications of membranes are widely used to separate or remove the unwanted components such as carbon dioxide, nitrogen, hydrogen sulfide and water vapor.

The focus in this project is gas separation by using membrane technology.

2.5 Types of Membrane in Gas Separation

Several factors that must be taken into account for material selection are reasonable cost, easy to handle, operating conditions and the most important is that the material can provide the best performance. In this case, the selectivity for CO_2 must also be high. Two areas of gas separation by using membrane research are organic (polymer) and inorganic membranes.[13]

2.5.1 Polymer Membrane

For polymer membrane, it is easy to be manufactured, however, it is only suited for low temperature application. The gas permeability and selectivity are determined by the polymer morphology and mobility. Figure 2.2 below shows an asymmetric hollow fiber membrane of functional cardo polyimide material supported by a porous structure which allows high permeability.

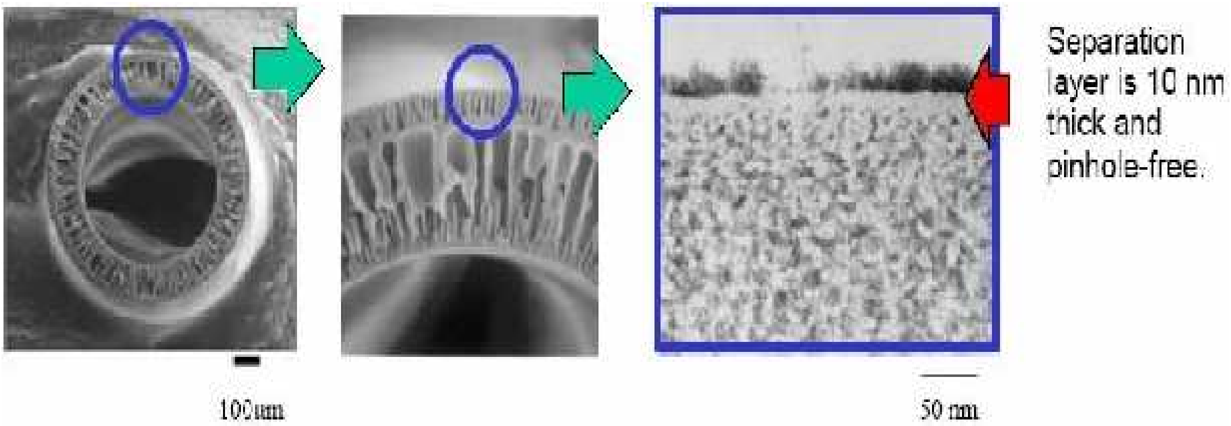


Figure 2.2: Cardo polyimide hollow fiber membrane with a thin, functional outer layer.

2.5.2 Inorganic Membrane

While for inorganic membrane, it has much greater thermal and chemical stability. Inorganic membranes can also withstand large pressure difference compared to polymer membranes.[14] The gas molecules are separated by effective size by sizing pores appropriately in materials including zeolites and silica which can act as molecular sieves, surface adsorption and diffusion inside the pores. Figure 2.3 illustrates gas separation using an ordered array of pores in an inorganic material. Selectivity can negatively affected by the defects in the pore structures.

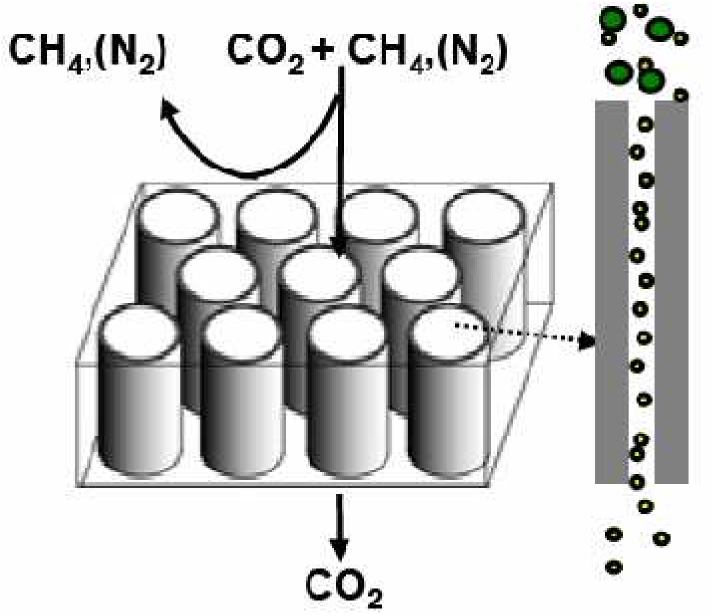


Figure 2.3: Porous inorganic membranes act as molecular sieves, differentiating gas molecules by effective size.

The limit of use of polymeric membrane is due to their poor performance stability at high pressure and temperature and in the presence of highly sorbing components. Besides that, high CO_2 partial pressures can plasticize polymer membranes, and thus decrease their separation ability.[15]

Comparison to microporous inorganic membrane, it has been studied for gas separation due to their superior thermal, mechanical and chemical stability, good erosion resistance, and high pressure stability. Several main types of microporous inorganic membranes are sol-gel derive ceramic membranes, chemical vapor deposition (CVD) modified membranes, leached hollow glass fibers, carbon molecular sieves and lastly zeolite membranes.

2.6 Transport Mechanism for Inorganic Membranes

In inorganic membrane, there are four main transport mechanisms by which gas separation using porous inorganic membranes can be described. The bases of these mechanisms are Knudsen diffusion, surface interactions (surface diffusion and capillary condensation), and the size of molecules (molecular sieving) to be separated.[15] The illustrations of listed transport mechanisms are shown below:

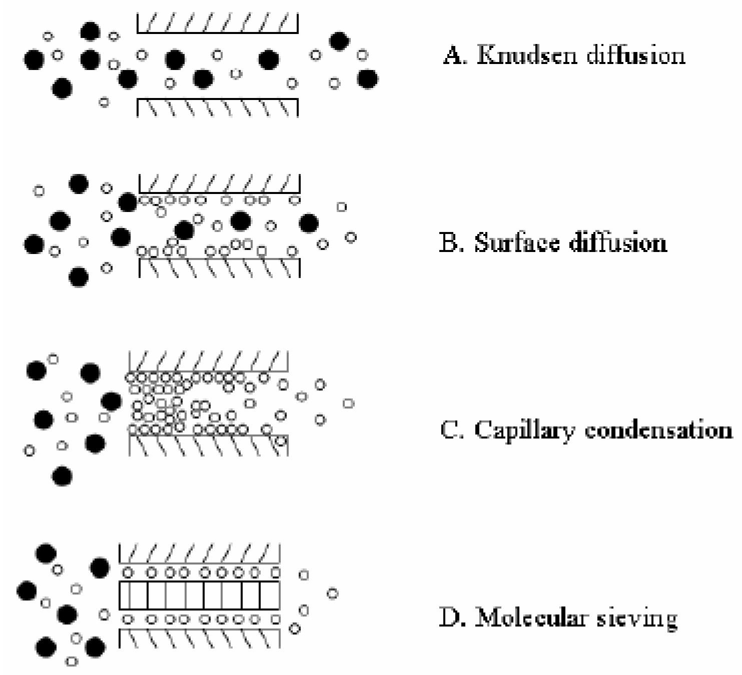


Figure 2.4: Transport mechanism through microporous membranes.

2.6.1 Knudsen Diffusion

Knudsen diffusion occurs when the mean free path of diffusing gas molecules surpasses the size of the pores through which diffusion proceeds.[16] It occurs in the gas phase through pores in the membrane layer which the diameter is smaller than the mean free path dimensions of the molecules in the gas mixture. The collisions are higher between the molecules with the pore wall compared with each other. Knudsen transport can occur by concentration or by pressure gradients. N_2 molecules preferentially permeate in the case of CO_2/N_2 separation. The selectivities of CO_2 with respect to N_2, CH_4, and H_2 by Knudsen diffusion will be 0.8, 0.6, and 4.7, respectively.[15] Hence, the selectivity of CO_2 achievable by the Knudsen mechanism is very low and not attractive in this particular gas mixture.

2.6.2 Surface Diffusion

In this mechanism, the diffusing species is adsorbed on the walls of the pore and then transport across the surface in the direction of surface concentration is decreasing. It is called as selective surface flow membranes, and several advantages has been mentioned for separation of gas mixtures using them.[17] Usually, the molecules with larger molecular weight, larger polarity and polarizability are selectively adsorbed on the membrane surface.[18] Temperature, pressure and nature of surface influence the concentration of adsorbed species.

2.6.3 Capillary Condensation

In capillary condensation, a pore is blocked by condensate, preventing gas transport of other components of the gas/vapor mixture.[15] Both aspects can result in increased selectivities. The condensation pressure depends on the pore size and shape and also the strength of the interaction between the fluid and pore walls.[15]

2.6.4 Molecular Sieving

In molecular sieving mechanism, separation happens when gas with smaller molecules pass through the porous membrane; while the other larger molecules cannot pass through the membrane because the kinematic diameter is larger than the pore size of membrane. High selectivity and permeability for the small gas molecules in a mixture can be obtained from molecular sieving membranes, but a very fine-tuning of the membrane pore sizes is required to achieve the desired separation efficiency.[17] Basically, this type of mechanism separates molecules based on the kinematic diameter.

Table 2.1: Molecular weight and kinematic diameter of CO_2, CH_4, N_2, and H_2**.**

	CO_2	CH_4	N_2	H_2
Molecular weight	44.01	16.04	28.01	2.02
Kinetic diameter, A	3.30	3.80	3.64	2.89

2.7 Alumina as Membrane Support

Alumina membranes had been used as a mesoporous membrane where separation takes place by a Knudsen diffusion mechanism.[19] However, the use of alumina membrane has become limited due to the selectivity in the region is limited and the rate of diffusion is controlled by molecular weight. For example, alumina membrane is not preferred in O_2/N_2 separation where the mass of the gases are about the same. It is because, O_2/N_2 is separated by diffusion, since the rate of diffusion for these molecules are about the same, and therefore, it is hard to separate it with the same diffusion. The limited use of alumina had also been approved in the experiments done by previous researchers.[20-21] Alumina has been used mainly as a support to membrane (such as silica membrane) because of its structural properties, and chemical and hydrothermal stabilities beyond 1000°C which makes it very desirable.[15]

2.8 Silica Membrane

Ceramic membrane is commonly made from alumina oxide (Al_2O_3), titanium oxide (TiO_2), zirconia oxide (ZrO_2), and silicon oxide (SiO_2).[22] In this thesis, alumina oxide disc is used as a support and the support is dipped into tetraorthosilicate (TEOS) solution where this solution provides silica sols from hydrolysis condensation reaction as a selective layer. In fabrication of CO_2 selective membranes, silica is usually considered as a viable starting material because of its stability.[15] The separation using silica membrane is mainly based on a molecular sieving effect where the molecules passing through the small pores based on their kinetic diameter.[23] Silica membrane with high combined values of flux and selectivity can be obtained through two methods of preparations which are sol-gel technique and chemical vapor deposition (CVD).

Microporous silica membrane is prepared by the deposition of a silica layer onto porous support.[15] The deposition of a silica layer can also be called as dip coating. The porous support can provide mechanical strength to the selective top layer silica. Since separation using silica membrane is mainly based on molecular sieving, the sol-gel technique is preferred because it can desirably control the pore size of the membrane.[22]

The sol-gel technique is divided by two which are colloidal route and polymer route. Colloidal sols membranes provide pore size in the ultra filtration range. It is used in separation of colloidal particles and large molecular weight solutes or used as a membrane support. As mentioned above, the separation in this project is based on molecular sieving effects and silica membranes with smaller than 1 nm pore size must be prepared.[22] Therefore, alumina membrane prepared in colloidal sol method is used as a support in the polymer sol route. The pore size in the polymer sol route is determined by the degree of branching of the inorganic polymer where a low degree of branching provides narrower pore system.[22] The sol-gel technique membranes provides high selectivities in gas separation.[22]

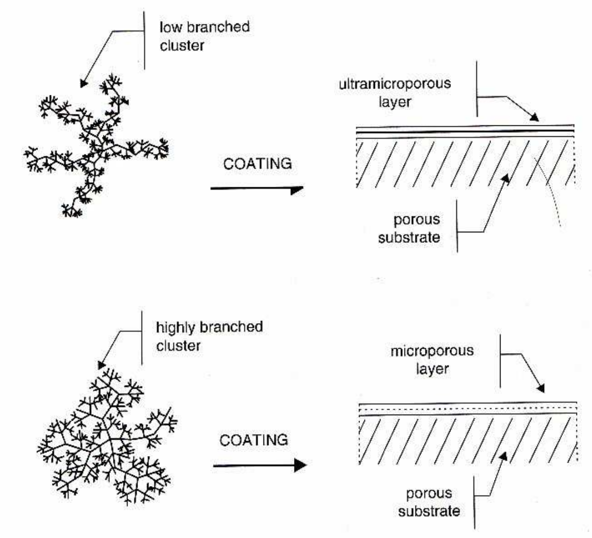


Figure 2.5: Low branch with narrower pores (above) and high amount branch with larger pores (below).

The silica pore size can be controlled by changing some parameters such as glass composition, annealing time, and temperature.[15] A study showed that an almost defect free silica membrane was prepared where two sol-gel silica layer was applied on top of the alumina support.[24] Beside that, the calcinations temperature used in the study also shows a different effect on the permselectivities. The CO_2/CH_4 permselectivities in the silica membrane which calcined at temperature of 400°C were very large and the H_2/CO_2 permselectivities were very large at temperature 600°C because the membranes were so dense that CH_4 could not permeate in membrane at 400°C and CO_2 in membrane at 600°C.[24]

In another study, it was reported that CO_2 permeance was increased through the silica membrane as the temperature decreased, while for N_2 and CH_4 permeances were increased very slightly. It is because CO_2 is more adsorptive on the silica surface than N_2 or CH_4.[25] Porous silica membranes were quite stable when it is used in dry conditions and a silica membrane on a silica-zirconia sub layer was even stable in humid conditions.[25] The other study found that the separation factors decreased as the temperature increased, which was due to the less permeable penetrant having larger activation energy.[26]

Table 2.2: Gas permeances for porous silica membranes.

Permeation Temperature (°C)	CO_2 Permeance (mol m^{-2} s^{-1} Pa^{-1})	Permselectivity		
		CO_2/N_2	CO_2/CH_4	H_2/CO_2
35	7×10^{-7}	17	80	2
300	2×10^{-7}	6	25	6

A study to analyze the effect of pressure has also been conducted. The study showed that the permeability for CO_2 was strongly increased with the mean pressure of the system in the other hand the permeability of N_2 and CH_4 were unchanged.[27] The transport through the silica modified membrane is due to the surface diffusion mechanism, as indicated by the observed pressure dependence and very small pores of the silica.[15] The permselectivities for CO_2/N_2 and CO_2/CH_4 were as high as 15 and 10, respectively, with CO_2 permeance of about 3 x 10^{-6} mol $m^{-2}s^{-1}$ Pa^{-1}.[15] In this project, inlet pressure, inlet flow rate and dip coating duration are varied to observe the effect on separation performance.

2.9 Fundamental Background

The driving force in membrane gas separation is the difference in partial pressure or concentration between the feed sides and permeate side.[28] Permeation rate, selectivity and flux are highly dependent with each other. Hence, the new membrane materials may combine different transport mechanism and thus increase the flux and selectivities.[28]

Transport occurs by a solution diffusion mechanism, and membrane selectivity is based on the relative permeation rates of the components through the membrane. Below is the general relationship in relating the permeation rate and flux. Diffusive flux through the membrane can be expressed by Fick's Law related to the membrane system as given: [28]

$$J_k = -D_k \frac{dC}{dz} \qquad (2.1)$$

Where: J_k : flux of component i (mole/m^2-s)
D_k : diffusivity of component i (m^2/s)
dC/dz: concentration gradient for component i over the length z (mol/(m^3m))

Fick’s law is integrated and applied for a membrane yields dx=*l* (membrane thickness), and dc_k=concentration difference (i.e. partial pressure for gases) over the membrane.

The permeance *P/l* [mol/(m^2.Pa.s)] is defined by

$$\frac{P_k}{l} = \frac{J_k}{\Delta p_k} \qquad (2.2)$$

P/l is referred to permeability flux and expressed as (m^3 (STP)/(m^2.bar.h)).

This equation shows that the flux through the membrane is proportional to the pressure difference across the membrane and inversely proportional to the membrane thickness**.**

From Equation 2.1,

$$J_k = -\frac{D_k}{RT}\left(\frac{dp}{dz}\right) \qquad (2.3)$$

It depends on the mean molecular speed, u and pore radius, r_p which is given by:

$$D_k = \frac{2}{3}ur_p \qquad (2.4)$$

An expression for the mean molecular speed can be obtained from the kinetic theory of gas:

$$u = \sqrt{\frac{8RT}{\pi M}} \qquad (2.5)$$

where

R and T are the gas constant and temperature respectively.

M is the molecular weight of the gaseous species ($g.mol^{-1}$).

Substituting equation (2.4) and (2.5) into equation (2.3) and then integrate it produces the permeation rate based on the Knudsen diffusion mechanism:

$$n_k = \left(\frac{32\pi}{9MRT}\right)^{1/2} r_p^3 \frac{p_h - p_h}{l} \qquad (2.6)$$

where n_k is the permeation rate is defined as:

$$n_k = \pi r_p^2 J_k \qquad (2.7)$$

Equation below shows the selectivity between gas components. The ideal separation factor, α could be expressed as the ratio of the pure gas permeabilities for the individual components i and j, for example, in this project, between CO_2 and CH_4.

$$\alpha_{CO_2/CH_4} = \frac{P_{CO_2}}{P_{CH_4}} \tag{2.8}$$

2.9.1 Flux and Permeability Calculation in the Project

In the permeability test experiment, the time (t) required to reach certain volume of gas in the permeate stream and volume of gas in the permeate stream (using a bubble soap flow meter) will be measured.[7] Flux and permeability can be calculated by using the equation below: [15]

The gas flux (J) through the membrane is defined by the following expression:

$$J = \frac{\Delta V}{At} \tag{2.9}$$

where ΔV is the volume of the permeated gas (cm^3)
A is the membrane area (cm^2)
t is the time (s)

The permeance (π) through the membrane is defined in equation (2.10) and is related to the permeability (P) by equation (2.11)

$$\pi = \frac{J}{\Delta p} \tag{2.10}$$

$$\pi = \frac{P}{L} \tag{2.11}$$

where Δp is the partial pressure difference between the upstream and downstream side of the membrane

L is the membrane thickness

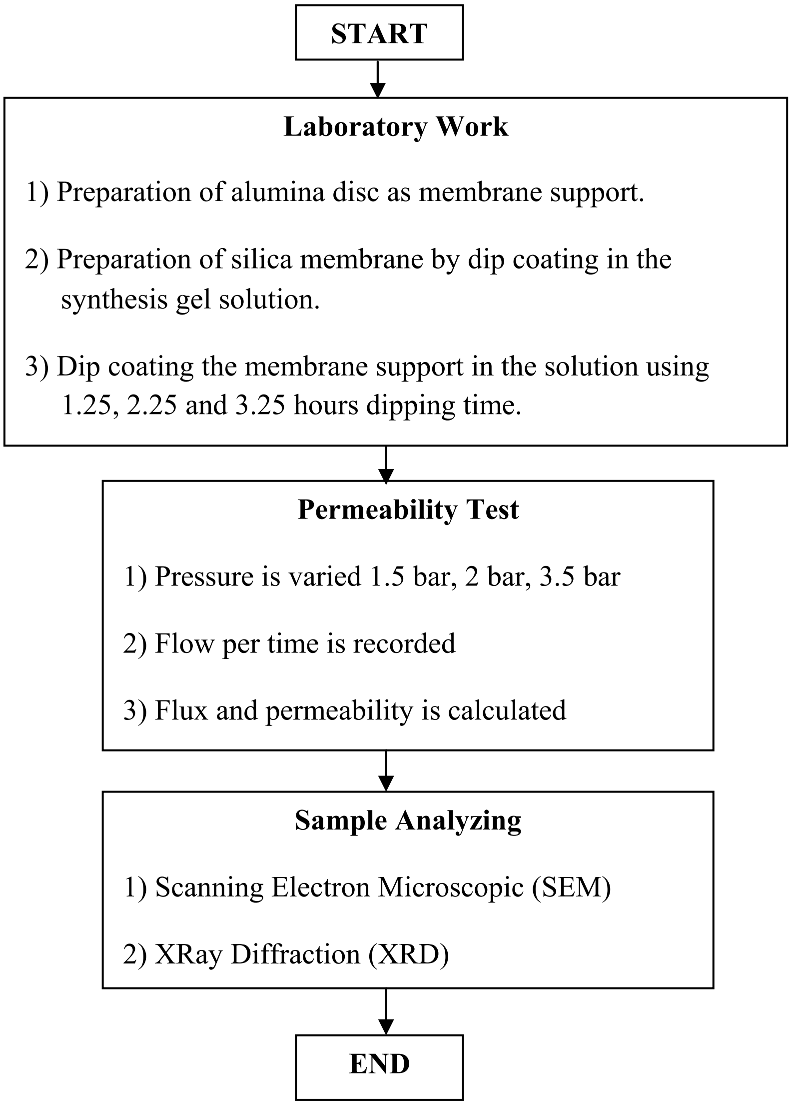
In this project, the research and study is on the performance of silica membrane which will be prepared by the sol-gel method. The experiment is done to analyze the performance of membrane in separating gas. Further discussion on methodology will be discussed in the next chapter.

‘

CHAPTER 3

METHODOLOGY

3.1 Research Methodology



START

Laboratory Work

1) Preparation of alumina disc as membrane support.
2) Preparation of silica membrane by dip coating in the synthesis gel solution.
3) Dip coating the membrane support in the solution using 1.25, 2.25 and 3.25 hours dipping time.

Permeability Test

1) Pressure is varied 1.5 bar, 2 bar, 3.5 bar
2) Flow per time is recorded
3) Flux and permeability is calculated

Sample Analyzing

1) Scanning Electron Microscopic (SEM)
2) XRay Diffraction (XRD)

END

Figure 3.1: Research methodology flowchart.

3.2 Project Gantt Chart

3.2.1 Gantt Chart for First Semester

No.	Detail/Week	1	2	3	4	5	6	7	Midsemester Break	8	9	10	11	12	13	14
1.	Project Topic Selection & Proposal Submission	■	■	■												
2.	Preliminary Research Work				■	■	■	■								
3.	Complete Chemical Form and Laboratory Work Form						■	■		●						
4.	Submission of Progress Report 1									●						
5.	Seminar 1															
6.	Project Work Continues										■	■	■	■	■	
	6.1 Chemical reservation															
	6.2 Familiarization with equipments for the project															
	6.3 Preparation of Silica Membrane															
																●
7.	Submission of Interim Report Final Draft															●
8.	Oral Presentation															

Table 3.1: Semester 1 Gantt Chart.

3.2.2 Gantt Chart for Second Semester

No.	Detail/Week	1	2	3	4	5	6	7		8	9	10	11	12	13	14
1.	Project Work Continue	■	■	■					Midsemester Break							
	1.1 Preparation of Silica Membrane															
						●										
2.	Submission of Progress Report 1															
3.	Project Work Continue				■	■	■	■		■	■	■	■			
	3.1 Preparation of Silica Membrane															
	3.2 Permeability Test,SEM, XRD															
										●						
4.	Submission of Progress Report 2															
5.	Seminar										■	■	■			
												●				
6.	Poster Exhibition															
														●		
7.	Submission of Dissertation (Soft bound)															
															●	
8.	Oral Presentation															
																●
9.	Submission of Project Dissertation (Hard bound)															

Table 3.2: Semester 2 Gantt Chart.

3.3 Chemicals and Tools Required

There are two experiments in this project. The chemicals and equipments/tools required are:

(i)For preparation of Alumina porous support

a) Aluminum Oxide
b) Starch

(ii)For preparation of Teraorthosilicate (TEOS) solution

a) Nitric Acid (HNO_3)
b) Ethanol (C_2H_5OH)
c) Tetraorthosilicate (TEOS)

(iii)Tools

a) Autopallet Machine
b) Furnace
c) Oven
d) Permeability Test Equipment

(iv)Characterization

a) Scanning Electron Microscopic (SEM)
b) X-Ray Diffraction (XRD)

3.4 Experimental Procedure

3.4.1 Preparation of Alumina Porous Support

This experimental procedure is the same as done by previous study.[7] Chemicals used in this experiment are aluminum oxide, starch and distilled water. 100 mL beaker is filled with distilled water and boiled it. Then, 15 gram of starch is added into beaker with boiled water. 10 gram of aluminum oxide is prepared on the glass plate. The dropper is used to mix solution of starch with alumina oxide and the number of starch drop is 10. The mixed powder is poured into a die and press using the autopallet machine. The same procedure is repeated with 20 drops of starch. Then, alumina support is heated at temperature 1300^{0}C in the furnace for 3 hours. Below is the picture of autopallet used to shape the alumina porous support.

Figure 3.2: Autopallet machine.

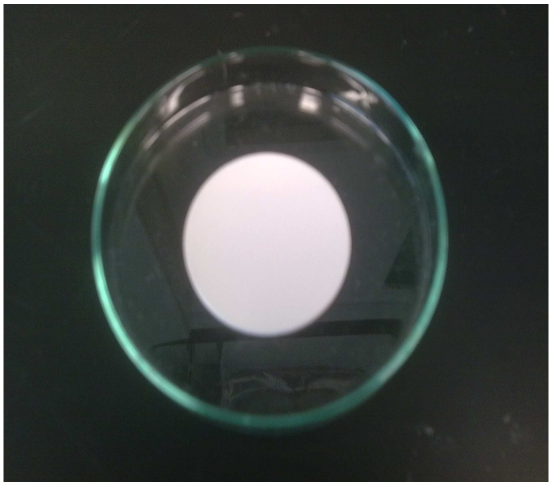


Figure 3.3: Alumina porous support.

3.4.2 Determination of porosity of Alumina Porous Support [29]

Alumina porous support is immersed in distilled water for 24 hours. After 24 hours, weight of wet alumina support is recorded. The equipment used for buoyancy weight is prepared as shown in Figure 3.4. After buoyancy weight is recorded, the alumina support is dried in oven at 100°C for 24 hours. Then, weight of alumina support after drying is recorded. Equation below is used to calculate the percentage of porosity.

$$\boxed{\%\ \text{Porosity} = \frac{\text{Weight of wet alumina disc} - \text{Weight of dried alumina disc}}{\text{Weight of wet alumina disc} - \text{Buoyancy weight of wet alumina disc}} \times 100\%}$$

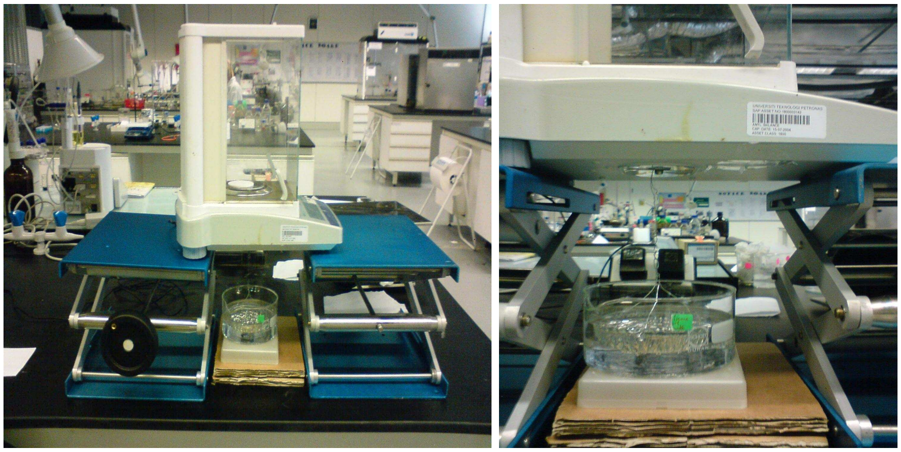


Figure 3.4: Equipment for buoyancy weight.

3.4.3 Preparation of Tetraorthosilicate (TEOS) Solution

10.63ml Tetraorthosilicate (TEOS), 53.15 Ethanol, 0.5ml Nitric Acid as catalyst, and 100ml distilled water are mixed in a beaker. The solution is left for 2 days.

Figure 3.5: Tetraorthosilicate (TEOS) solution prepared.

3.4.4 Dip Coating [30]

This method is used to combine the alumina support and silica solution to prepare a silica layer (dense layer) on the surface of alumina support. The dip coating hours are varied to observe the effect of permeability and selectivity on the thickness of membrane. The alumina support is immersed in the TEOS solution for 1.25, 2.25 and 3.25 hours (Note: Only one side of alumina support surface must be dipped in the solution). Alumina support with coated silica layer is dried at 100^{o}C for 2 hours. Lastly, alumina support with coated silica layer is sintered in furnace at 500^{o}C for 4 hours.

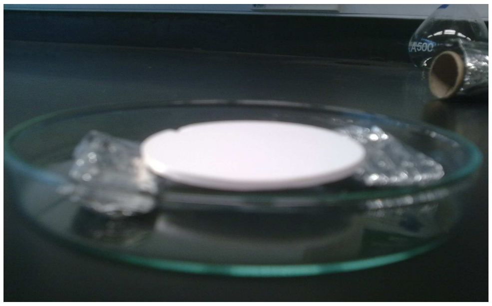


Figure 3.6: Dip coating.

3.4.5 Permeability Test Equipment

The permeability test equipment is available at Membrane Fabrication Unit (MFU) laboratory. It is used to measure the gas permeation through the membrane fabricated. This equipment consists of a feed gas tank, a pressure gauge of inlet gas, a dead-end membrane cell where membrane is located and a bubble soap flow meter to measure the volume of gas in permeate stream. In this test, the time required to reach certain volume of gas in the permeate stream is observed and recorded. Since the membrane separation is driven by pressure difference, the pressure drop through the experiment is varied to see the effect on permeability. In this experiment, the volume of permeate gas is fixed which is 50 cm^3 and the time taken for the soap bubble to reach 50 cm^3 in the pipette is recorded. The inlet pressure is varied for 1.5 bar, 2.0 bar, and 3.5 bar. The permeability equipment is shown below.

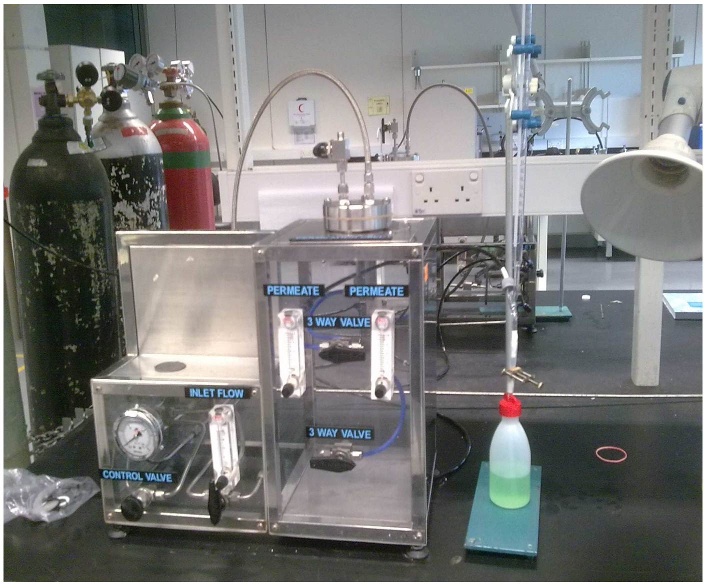


Figure 3.7: Permeability test equipment.

Below is the schematic diagram of permeability test equipment.

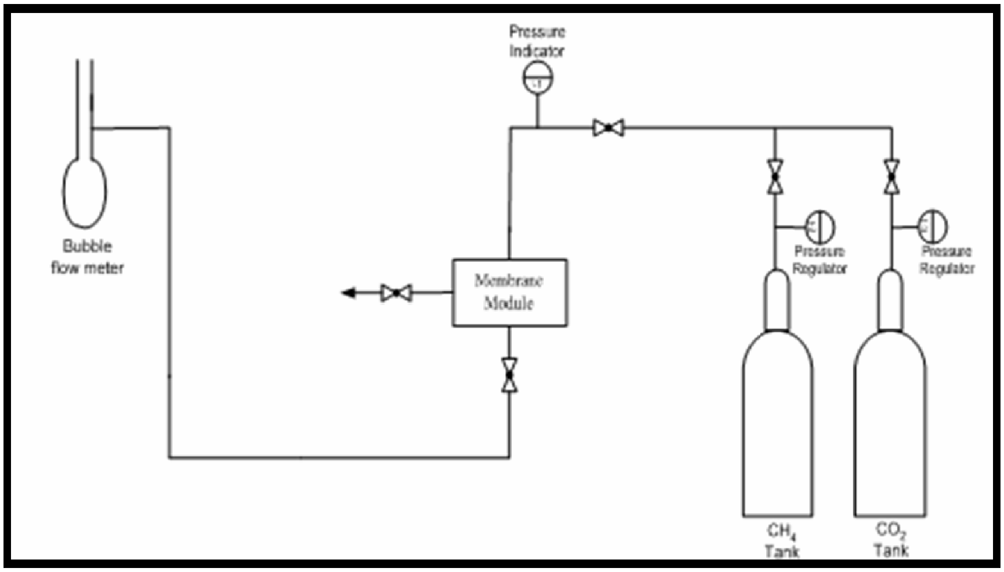


Figure 3.8: Schematic diagram of permeability test.

3.4.6 Scanning Electron Microscopic (SEM)

SEM is used to analyze the condition and structure of membrane surface and the cross section of the membrane. Cross section characterization is needed to analyze the thickness of the silica membrane coated on the membrane after the dip coating. Below is the picture of SEM equipment.

Figure 3.9: Scanning Electron Microscopic (SEM).

CHAPTER 4

RESULT & DISCUSSION

4.1 Porosity for Alumina Porous Support

In the preparation of Alumina support, the percentage of porosity of Alumina support is calculated. The diameter of Alumina support is 5 cm and the thickness is 0.3 cm. The data taken for porosity calculation is shown below:

Table 4.1: Data recorded for porosity calculation.

Sample	No. of Starch (drop)	Weight in air(g)	Weight of buoyancy(g)	Weight in water for 24 hours (g)	Porosity (%)
1	10	9.831	3.21	11.383	18.99
2	20	9.802	3.39	11.599	21.89

As shown in Table 4.1, as the number of starch used is increased from 10 to 20 drops, the percentage of porosity is also increases from 18.99% to 21.89%. This is because, when starch is mixed, it can cause the disruption to alumina particles, wall effect and also the contact effect.[31] The interconnection derived between the pores will result in higher porosity. Therefore, when the amount of starch added is increased, the porosity is also increases.

4.2 Permeability Test Result

4.2.1 Flux Data

The permeability test was done by varying the inlet pressure (1.5 bar, 2.0 bar, 3.5 bar) and for each pressure, the inlet flow used was 1 l/min (16.67 cm^3/s), 2 l/min (33.33 cm^3/s), and 3 l/min (50.00 cm^3/s). Table 4.2 shows the flux calculated for 1.25, 2.25, and 3.25 hours dip coating duration for different inlet pressure and inlet flow rate.

Table 4.2: Data of flux calculated.

Inlet Pressure (bar)	Inlet Flow (cm^3/s)	Flux,J (cm^3[STP] / cm^2.s)					
		CO_2 (1.25h)	CH_4 (1.25h)	CO_2 (2.25h)	CH_4 (2.25h)	CO_2 (3.25h)	CH_4 (3.25h)
1.5	16.67	0.7410	0.7311	0.7369	0.7180	0.7300	0.7078
1.5	33.33	1.0034	0.9877	0.9736	0.9199	0.9696	0.8496
1.5	50.00	1.5763	1.4211	1.4863	1.4374	1.6828	1.3309
2.0	16.67	0.7713	0.7544	0.7593	0.7408	0.7519	0.7423
2.0	33.33	1.0731	1.1444	1.0527	1.1383	1.1514	1.1024
2.0	50.00	1.8544	1.6134	1.8136	1.5218	1.7328	1.3945
3.5	16.67	0.7867	0.7804	0.7830	0.7804	0.7710	0.7617
3.5	33.33	1.3045	1.2111	1.2049	1.1745	1.1926	1.1745
3.5	50.00	1.9110	1.6111	1.8089	1.5910	1.7904	1.4404

Below is the graph for Flux versus Inlet Flow for different inlet pressure.

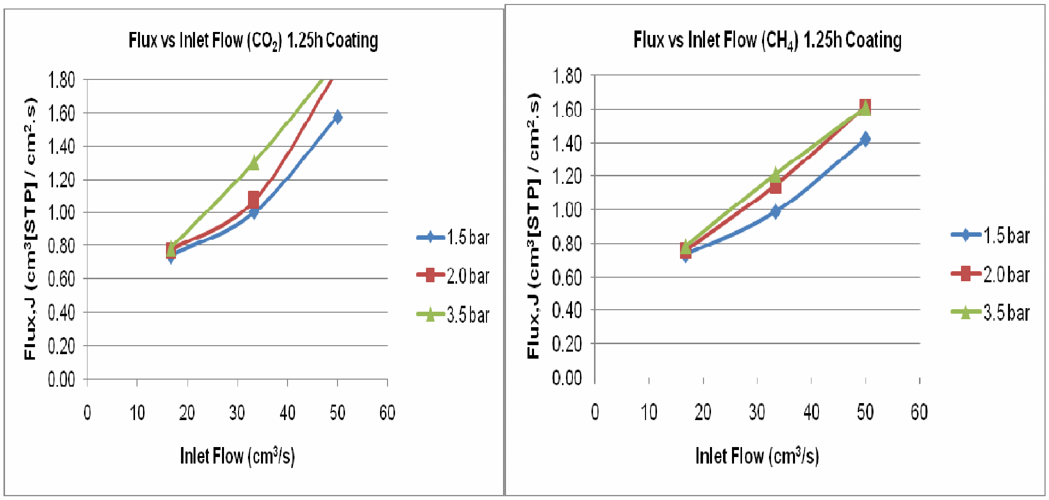


Figure 4.1: Graph of Flux vs. Inlet Flow for 1.25 hours dip coating.

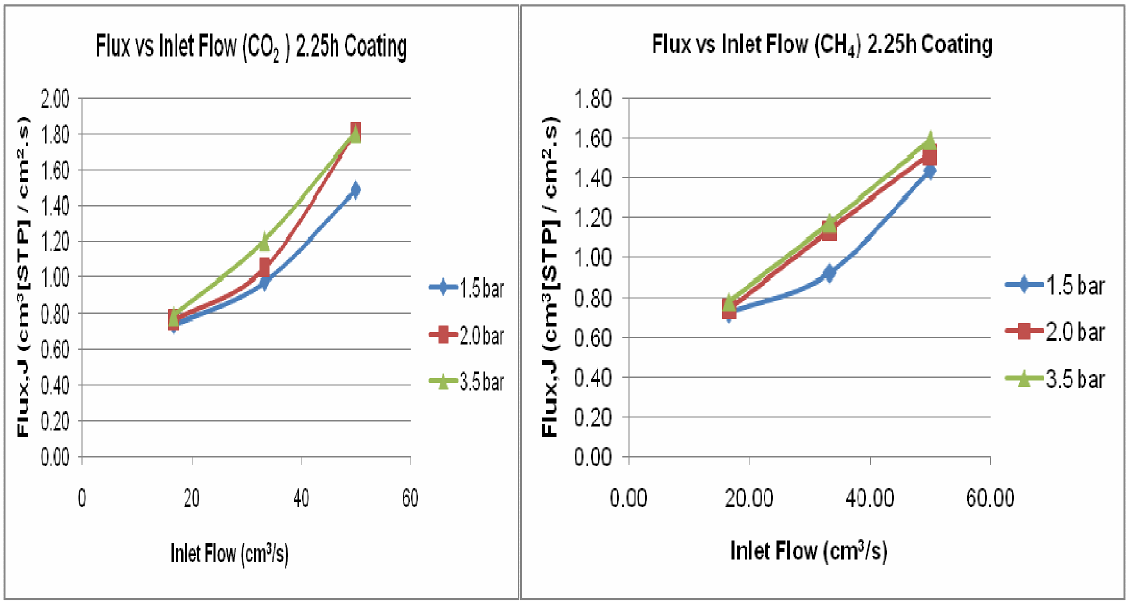


Figure 4.2: Graph of Flux vs. Inlet Flow for 2.25 hours dip coating.

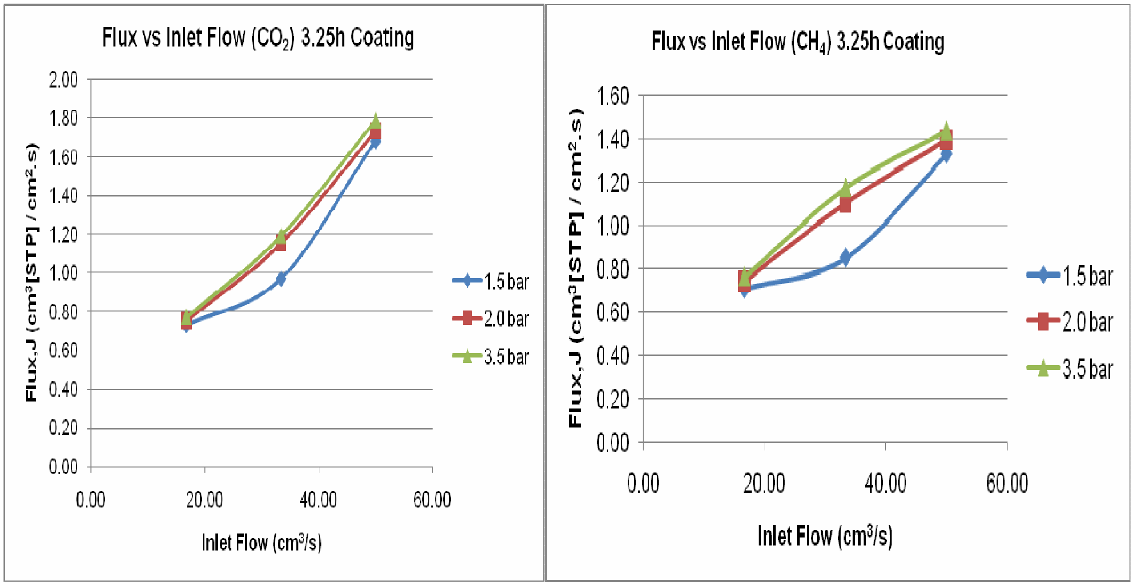


Figure 4.3: Graph of Flux vs. Inlet Flow for 3.25 hours dip coating.

Figure 4.1, 4.2 and 4.3 are the graph of Flux vs. Inlet flow. It shows that, when the inlet flow or feed flow is increased, the flux is increases. The higher the inlet flow rate, the easier for the gas molecules to pass through the membrane resulting in higher flux. CO_2 has higher adsorption coverage towards silica layer making more CO_2 diffuses through the membrane.[4] Thus, this will reduce the time taken for the CO_2 to reach volume of 50 cm^3 in the permeability test. However, the flux for CO_2 is higher than CH_4. This is due to smaller size of CO_2 (3.30A) compared to CH_4 (3.38A). Besides that, in the surface diffusion effect, the molecules with larger molecular weight (CO_2=44.01 g/mol and CH_4=16.04 g/mol) are selectively adsorbed on the membrane surface.[18] Higher flux for CO_2 is also because of its linear molecular shape compared to CH_4 with tetrahedral shape which makes CH_4 molecules bigger to pass through the membrane. The graph below shows the flux for CO_2 and CH_4 and each graph is plotted for different dip coating duration which is 1.25 hours, 2.25 hours and 3.25 hours.

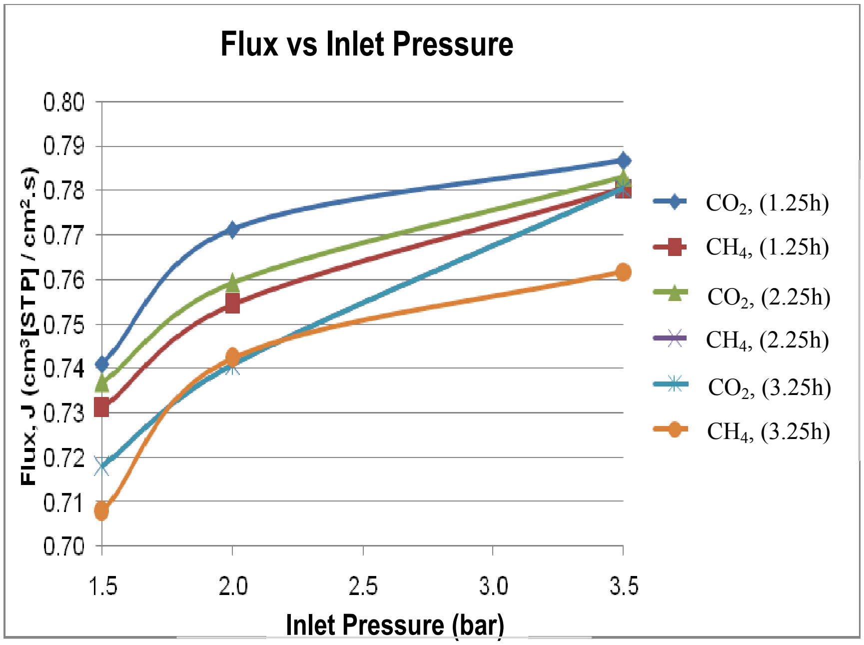


Figure 4.4: Graph of Flux vs. Inlet Pressure for 16.67 cm^3/s Inlet Flow Rate.

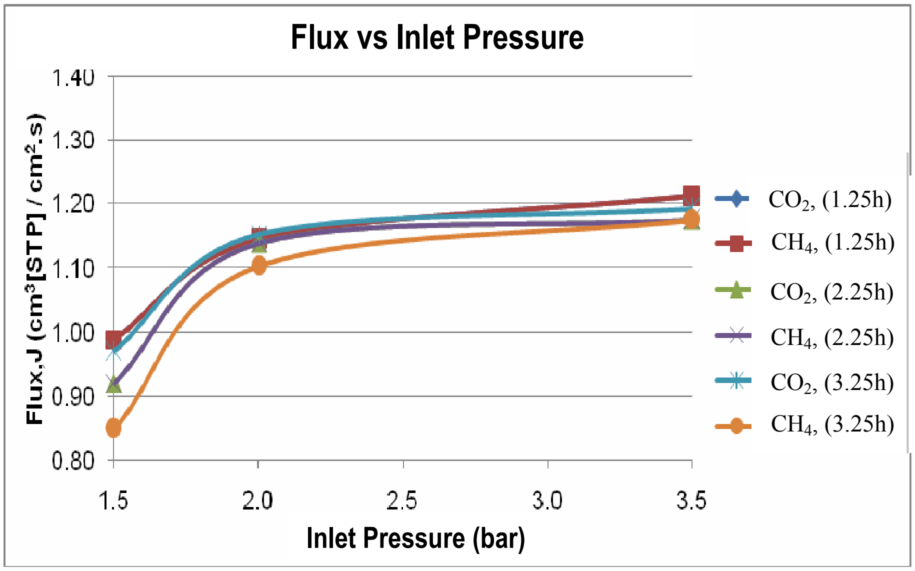


Figure 4.5: Graph of Flux vs. Inlet Pressure for 33.33 cm^3/s Inlet Flow Rate.

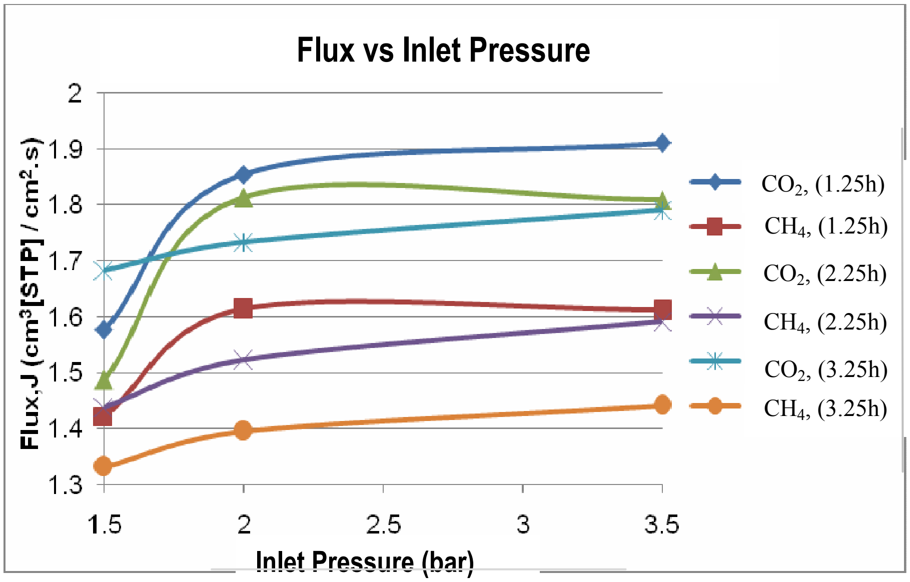


Figure 4.6: Graph of Flux vs. Inlet Pressure for 50.00 cm^3/s Inlet Flow Rate.

As shown in Figure 4.4, 4.5 and 4.6, flux increases when the inlet pressure is increased. When the partial difference increased, it increases the gradient of adsorbed concentration, the flux increases.[4] Figure 4.4, 4.5 and 4.6 also shows the effect of flux on different duration of dip coating hours. Dip coating duration used is 1.25, 2.25 and 3.25

hours. As can be seen in the figure above, it shows that the flux for 1.25 hour dip coating is higher than the flux for 2.25 hours dip coating followed by 3.25 hours dip coating. Scanning Electron Microscopic was done to get the silica layer thickness. However, SEM only managed to observe the thickness of 2.25 and 3.25 hours dip coating only. 3.25 hours dip coating results in thicker silica layer which is 289.8 μm compared to 2.25 hours dip coating with 88.20 μm thickness.

Silica layer acts as a selective layer in membrane separation. The gas molecules are adsorbed to silica layer first then passes through the porous according to pressure and concentration difference. Therefore, the thicker silica layer is, more molecules will be absorbed first, and then only it passes through the porous of membrane. This is the reason of having larger flux for membrane with 2.25 hours dip coating compared to 3.25 hours dip coating.

4.2.2 Permeability Data

Table 4.3 below shows the data of permeability calculated.

Table 4.3: Data of permeability calculated.

P_{inlet} (bar)	Inlet Flow (cm^3/s)	Permeability (cm^3[STP].cm/cm^2.bar.s)					
		CO_2 (1.25h)	CH_4 (1.25h)	CO_2 (2.25h)	CH_4 (2.25h)	CO_2 (3.25h)	CH_4 (3.25h)
1.5	16.67	0.444600	0.438660	0.442125	0.430788	0.437976	0.424690
1.5	33.33	0.602040	0.592620	0.584171	0.551930	0.581743	0.509731
1.5	50.00	0.945780	0.852660	0.891759	0.862461	1.009660	0.710415
2.0	16.67	0.231390	0.226320	0.227776	0.222232	0.225574	0.222703
2.0	33.33	0.321930	0.343320	0.315804	0.341479	0.345410	0.330723
2.0	50.00	0.556320	0.484020	0.544066	0.456542	0.519825	0.400345
3.5	16.67	0.094404	0.093648	0.093964	0.093650	0.092515	0.091408
3.5	33.33	0.156540	0.145332	0.144585	0.140946	0.143107	0.140946
3.5	50.00	0.229320	0.193332	0.217064	0.190918	0.214843	0.172847

Below are the graphs of Permeability vs. Inlet Pressure for different inlet flow rate and different coating hours.

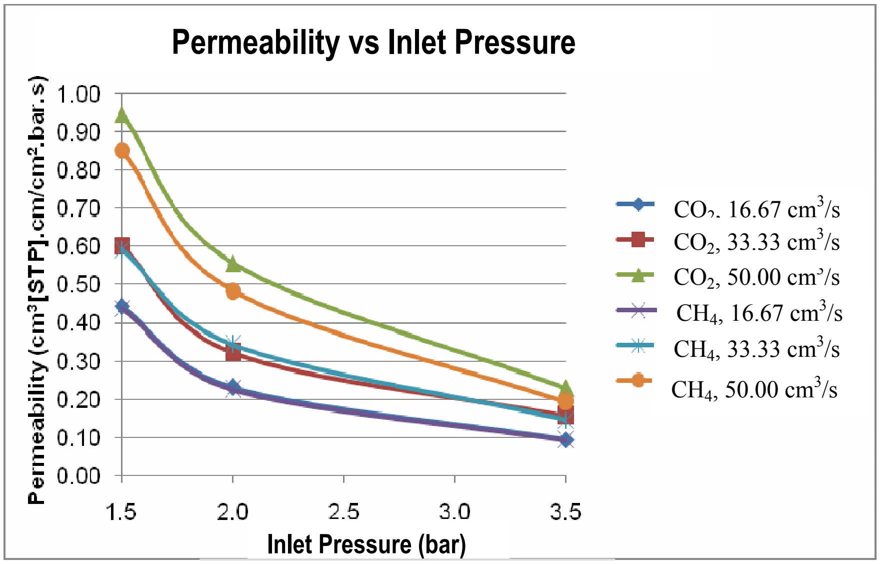


Figure 4.7: Graph of Permeability vs. Inlet Pressure for 1.25 hours dip coating.

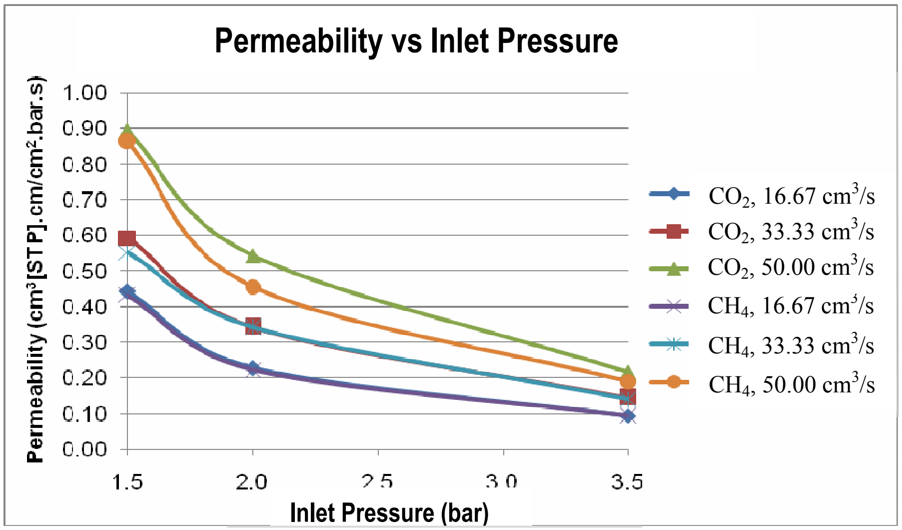


Figure 4.8: Graph of Permeability vs. Inlet Pressure for 2.25 hours dip coating.

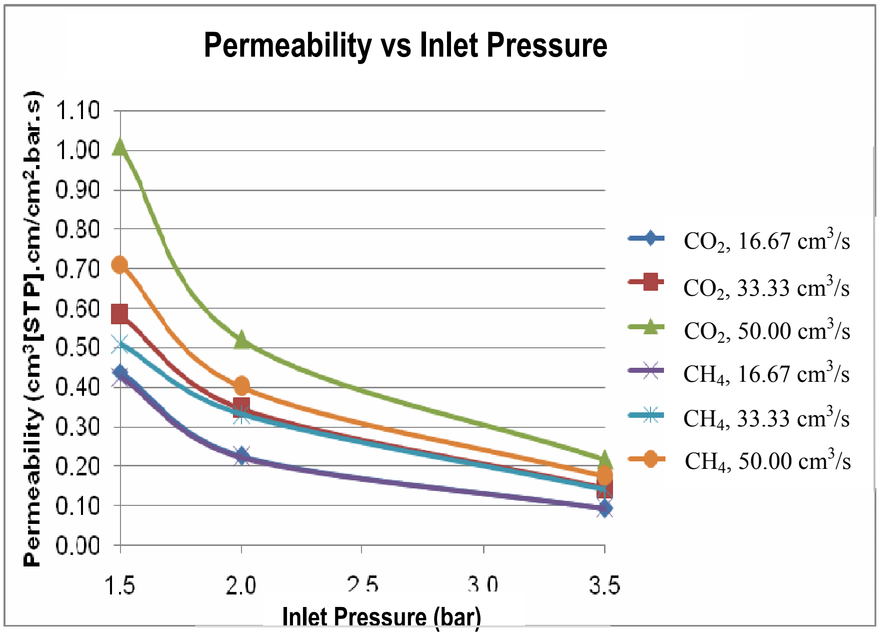


Figure 4.9: Graph of Permeability vs. Inlet Pressure for 3.25 hours dip coating.

Based on Figure 4.7, 4.8, and 4.9, it shows that when inlet pressure is increased, the permeability of gases decreases. As inlet pressure is increased, the driving force is increased and more molecules pass through the membrane. However, as pressure increased, more molecules trying to pass through the membrane making the molecules concentration in the inlet side increases and approaches the saturation thus reducing the permeability of CO_2 and CH_4.[4] Highest permeability was for 3.25 hours dip coating followed by 2.25 hours and 1.25 hours dip coating is because of the silica layer thickness. Silica layer acts as selective layer, although the process is slow in flux for 3.25 hours dip coating, but, it can adsorb more molecules compared to thinner silica layer, thus, more amounts of molecules can pass through the membrane resulting in higher permeability.

Below is the separation factor for this thesis.

Table 4.4: Separation factor, α.

Inlet Pressure (bar)	Inlet Flow (cm^3/s)	Separation Factor,α		
		CO_2/CH_4 1.25h Coating	CO_2/CH_4 2.25h Coating	CO_2/CH_4 3.25h Coating
1.5	16.67	1.0135	1.0263	1.0313
1.5	33.33	1.0159	1.0584	1.1413
1.5	50.00	1.1092	1.0340	1.4212
2.0	16.67	1.0224	1.0249	1.0129
2.0	33.33	0.9377	0.9248	1.0444
2.0	50.00	1.1494	1.1917	1.2984
3.5	16.67	1.0081	1.0034	1.0121
3.5	33.33	1.0771	1.0258	1.0153
3.5	50.00	1.1861	1.1370	1.2430

As shown in Table 4.4, the highest separation factor is 1.4212 at 1.5 bar for 50.00cm^3/s inlet flow for 3.25 hours dip coating compared to previous thesis with the highest separation factor is 1.37 which is lower.[7] Separation factor shows the ability of separating CO_2 and CH_4, thus, high separation factor must be obtained to ensure the efficiency of the separation.

4.3 Scanning Electron Microscopic (SEM) Result

From the Scanning Electron Microscopic (SEM), Figure 4.10 shows the thickness of the silica layer for 2.25 hours of dip coating which is 88.20 µm and Figure 4.11 shows for 3.25 hours dip coating which is 289.8 µm. The longer dip coating duration is, the thicker silica layer will be formed. Comparing to previous thesis, the writer managed to get the thickness 9µm for 1.25 hours dip coating, 13.9 µm for 2.25 hours dip coating and 15 µm for 3.25 hours dip coating.[7] Based on the values, this thesis managed to get larger thickness of silica layer compared to previous thesis.

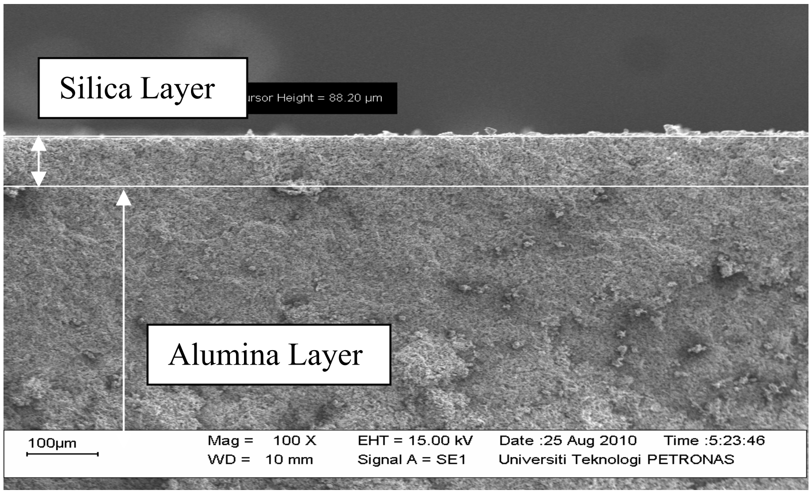


Figure 4.10: SEM (100X) image for 2.25 hours dip coating.

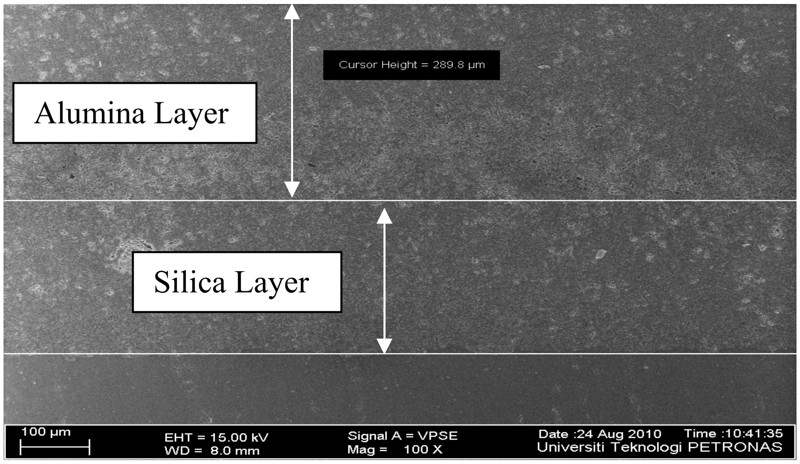


Figure 4.11: SEM (100X) image for 3.25 hours dip coating.

Figure 4.12 below is the image of 2.25 hours dip coating at 30X magnification which the silica layer (above) is clearer to be seen.

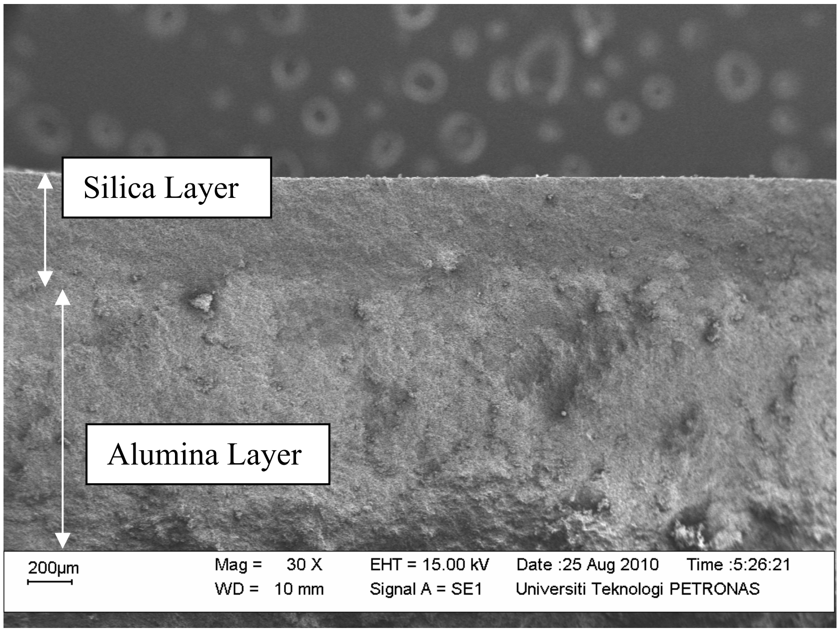


Figure 4.12: SEM (30X) image for 2.25 hours dip coating.

4.4 X-Ray Diffraction (XRD) Result

Below are the graph results from the XRD for three different coating hours. Figure 4.15 for 3.25 hours dip coating duration shows highest peak followed by Figure 4.14 for 2.25 hours and Figure 4.13 for 1.25 hours dip coating duration. It shows that, the longer the dip coating duration is, more silica are deposited onto the porous alumina surface resulting in thicker silica layer.

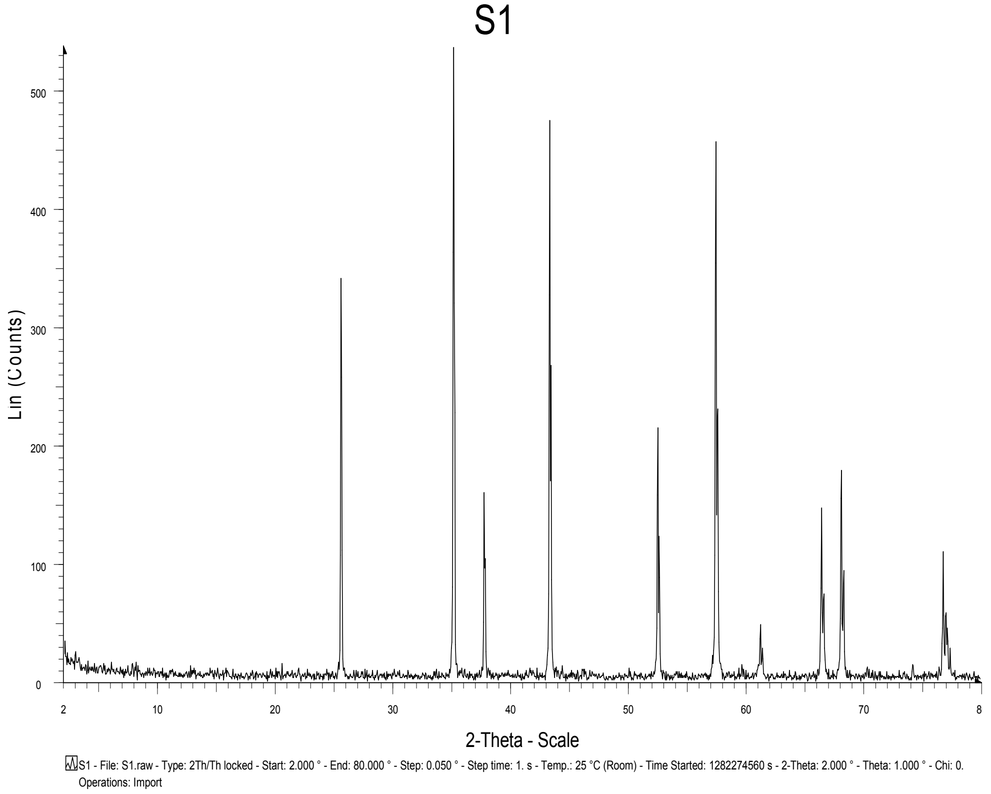


Figure 4.13: XRD graph for 1.25 hours dip coating.

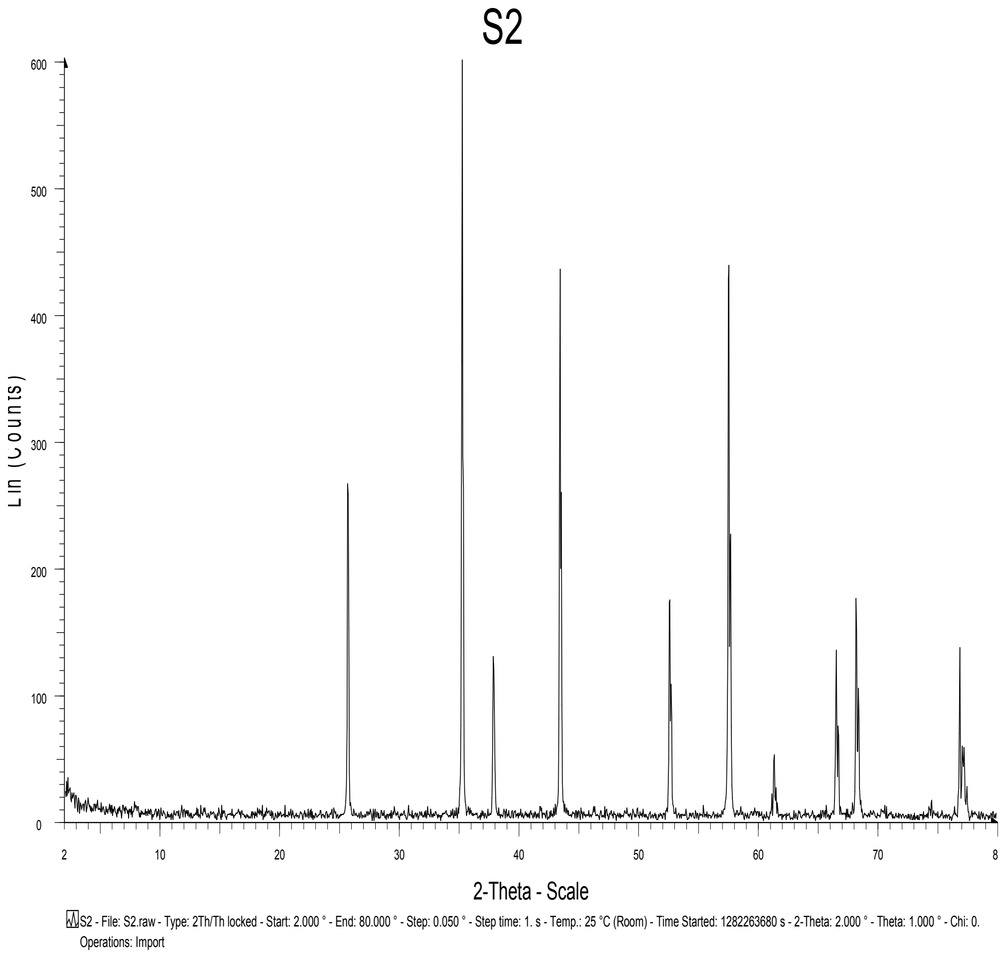


Figure 4.14: XRD graph for 2.25 hours dip coating.

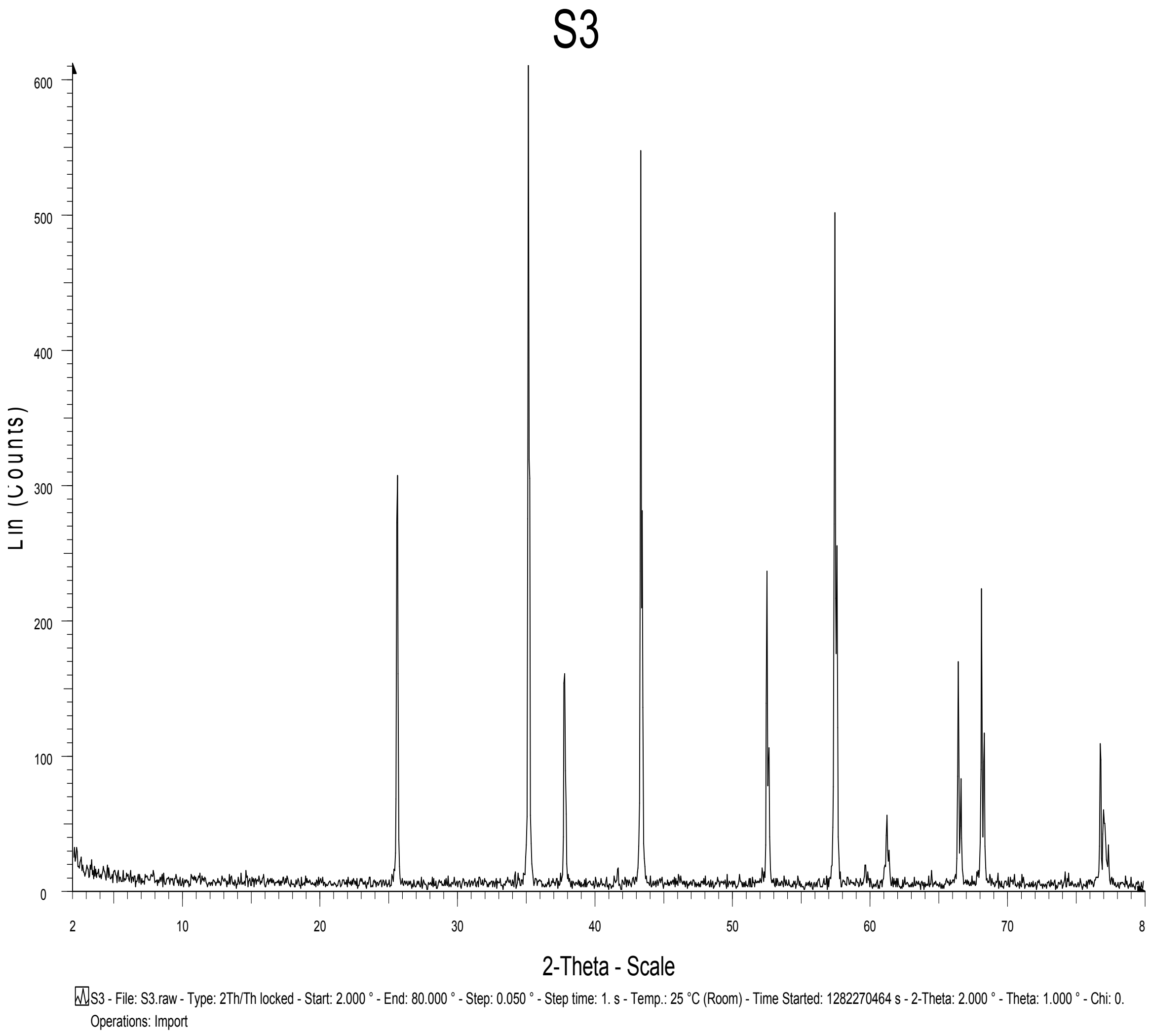


Figure 4.15: XRD graph for 3.25 hours dip coating.

CHAPTER 5

CONCLUSION & RECOMMENDATION

In this project, silica membrane is prepared through the deposition of silica sol onto alumina porous support. Porosity percentage increases from 18.99% to 21.89% when starch drop is increased from 10 drops to 20%. It shows that starch can be used as a binder to control the porosity of alumina support. As shown in Table 4.2, flux is also increases when the inlet flow rate and inlet pressure are increased. The highest flux is 1.911 cm^3[STP]/cm^2.s at 3.5 bar for 50.00 cm^3/s inlet flow rate. However, flux decreases as the dip coating hours is increased (also when the silica thickness increases). In the other hand, flux for CO_2 is higher compared to CH_4 due to adsorption and diffusion effect which depends on the molecular size of the gas molecules.

The permeability decreases as the inlet pressure is increased due to concentration of gas molecules approaches saturation in the inlet side. However, permeability increases as the dip coating hour is increased results in more molecules adsorbed onto the silica layer. The highest permeability is for CO_2 which is 1.009660 cm^3 [STP].cm/cm^2.bar.s at 1.5 bar for 50.00 cm^3/s inlet flow rate at 3 hours dip coating duration. The separation factor is the highest at 1.5 bar for 50.00 cm^3/s inlet flow rate at 3 hours dip coating which is 1.4212. Thus, it can be concluded that as inlet flow rate is increased, flux is increases. For permeability, as the inlet pressure is increased, the pressure drop is increases, the flux increases and permeability decreases.

Although this experiment did not achieve the ideal separation factor which is 1.66, but this project is able to increase the separation factor, 1.4212 which is slightly higher than previous thesis with separation factor 1.37.

During conducting the experiment, there might be some errors occur as follow:

1) Human error in reading and recording the time taken during the permeability test as the soap bubbles move very fast. The time recorded might not be accurate.
2) The membrane used in permeability test might have some cracks which cannot be seen directly.

There are a few recommendations can be made in the experiment:

1) Install a digital time reader or flow rate reader into the permeability equipment.
2) Take more reading and find the average.
3) Use mix gas of CO_2 and CH_4 to get the more accurate results.
4) The researcher can add one more parameter which is checking the hardness of the membrane.
5) Multilayer of selective layer can be used to increase the efficiency of membrane.

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APPENDIX

A) Raw Data

Below is the data taken for the gas to reach 50 cm^3 in the permeability test.

Inlet Pressure (bar)	Inlet Flow (cm^3/s)	Flux,J (cm^3[STP] / cm^2.s)					
		CO_2 (1.25h)	CH_4 (1.25h)	CO_2 (2.25h)	CH_4 (2.25h)	CO_2 (3.25h)	CH_4 (3.25h)
1.5	16.67	3.44	3.48	3.46	3.55	3.49	3.60
1.5	33.33	2.54	2.58	2.62	2.77	2.63	3.00
1.5	50.00	1.62	1.79	1.71	1.77	1.51	1.91
2.0	16.67	3.30	3.38	3.35	3.44	3.39	3.43
2.0	33.33	2.37	2.23	2.42	2.24	2.21	2.31
2.0	50.00	1.37	1.58	1.40	1.67	1.47	1.83
3.5	16.67	3.24	3.26	3.25	3.26	3.30	3.34
3.5	33.33	1.95	2.10	2.11	2.17	2.14	2.17
3.5	50.00	1.33	1.58	1.41	1.60	1.42	1.77

B) Example of calculation for percentage of porosity

Below is the data taken for porosity percentage calculation.

No. of Starch (drop)	Weight in air(g)	Weight of buoyancy(g)	Weight in water for 24 hours (g)	Porosity (%)
20	9.802	3.39	11.599	21.89

The formula used to calculate percentage of porosity is:

$$\% \text{ Porosity} = \frac{\text{Weight of wet alumina disc} - \text{Weight of dried alumina disc}}{\text{Weight of wet alumina disc} - \text{Buoyancy weight of wet alumina disc}} \times 100$$

$$= \frac{(11.599 - 9.802)}{(11.599 - 3.39)} \times 100$$

$$= 21.89\ \%$$

C) Example of calculation for permeability

Data for 1.5 bar (16.67 cm^3/s) and 3.25 hours dip coating for CH_4.

1) Convert Q to Q_{STP}

$$Q_{STP} = (T_{STP}/T_{ROOM}) \text{ x } (V/t)$$

$$= (273K/295K) \text{ x } (50 \text{ cm}^3 / 3.30 \text{ s})$$

$$= 13.89 \text{ cm}^3[STP] / s$$

2) Flux, J

$$J = Q_{STP}/Area$$

$$= (13.89 \text{ cm}^3[STP] / s) \; / \; (19.63 \text{ cm}^2)$$

$$= 0.7078 \text{ cm}^3[STP] / \text{cm}^2.s$$

3) Permeability, P

$$P = (\text{ Flux / Pressure Difference}) \text{ x Membrane Thickness}$$

$$= (J / \Delta p) \text{ x } L$$

$$= \frac{(0.7078 \text{ cm}^3[STP] / \text{cm}^2.s) \text{ x } (0.3 \text{ cm})}{0.5 \text{ bar}}$$

$$= 0.42469 \text{ cm}^3[STP].\text{cm} / \text{cm}^2.\text{bar.s}$$