

# **Gas Separation using Acrylonitrile Butadiene Styrene Membrane**

Dissertation

By

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CERTIFICATION OF APPROVAL

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BACHELOR OF ENGINEERING (Hons)

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Approved by,

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(Associate Professor Dr. Mohamad Azmi Bustam)

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.

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(Mohd.Khairul Anuwar Bin Hussin)

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## **ABSTRACT**

This report is discussing mainly about the permeation of gases using Acrylonitrile Butadiene Styrene (ABS) membrane. ABS is chosen because its cost and properties that made it suitable for gas separation. In this project, focus is given fabrication of the said membrane and permeation experiment on the membrane. The gases which are used for the study is CH<sub>4</sub>, O<sub>2</sub> and N<sub>2</sub>. Two types of membrane are fabricated in the studies which are porous and dense. Solution-casting technique is used to fabricate dense while porous membrane is fabricated by phase inversion (solution precipitation). The membranes are tested for its permeability for the three gases using Gas Permeability Test Unit. Based on the result, dense membranes have better ideal selectivity compared to the porous membrane. The result obtained will be useful in determining the potential of ABS as membrane for gas separation.

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# CHAPTER 1

## INTRODUCTION

Natural gas is widely used all over the world as the combustion fuel after crude oil and its products. Natural gas contains light hydrocarbon mainly methane with some amount of ethane, propane, butane and pentane as well as considerable amount of contaminants or non-hydrocarbons such as nitrogen, carbon dioxide, hydrogen sulfide, water vapour and also trace amount of mercury and helium.

The presence of nitrogen reduces the quality of natural gas in a sense that it lowers the heating value of natural gas which then reducing the heat (energy) produced during the combustion of the gas. In addition, excess amount of nitrogen in natural gas makes it unsuitable for pipeline transportation as the limit is four mol% of nitrogen maximum.

The Cryogenic Distillation process has been commercially used worldwide for removal of nitrogen from natural gas in various Gas Processing Plants but this technique requires high capital and operating costs. Furthermore, Cryogenic process also associated with mechanical and operational complexity. Separation of N<sub>2</sub> and O<sub>2</sub> from is also important in industry. The separation process is used to produce feed gases that will be used for chemical production. The process of separating these gases is also dependent on Cryogenic Distillation.

To solve this problem, “membrane technology has been widely developed. In order to ensure the practicality of the membrane application, several factors have to be observed, including economical, easily scalable for industrial application and having sufficient permeability and selectivity” (Porter, 1990). Thanks to the vigorous study in the field of membrane sciences, significant advancement in membrane technology has been made in recent years.

There are many alternatives that can be selected when dealing with organic membranes. One of the popular ones is polycarbonate and its variation including

mixed matrix membrane. With aggressive research, its property regarding its characteristic such as permeability, for CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and O<sub>2</sub> is well documented (Chen et al, 2000), (Şen et al, 2007).

Polyamide membrane is an interesting high performance polymer for membrane fabrication because of its thermal stability and chemical resistance. (Chern and Huang, 1998). This property can be improved by introducing another homopolymer to form copolymer. It is an alternative to obtain better membrane material other than creating a new homopolymer. For example, polyamides membrane is shown to have increased selectivity by cross-linking it with another homopolymer, which is 4, 4'-diaminobenzoylanilide (DABA) using butylene glycol (Kim et al, 2006)

## **1. Problem statement**

Organic membranes based on Acrylonitrile Butadiene Styrene (ABS) copolymer can be used for gas separation. Its performance can be measured from its permeability and selectivity. Porous membranes usually have high permeability and low selectivity and dense membrane is vice-versa. The ideal membrane for separation process should have high selectivity while still maintaining reasonable permeability. This project is assessing the possibility of using ABS copolymer resin trademark Toyolac for dense and porous membrane fabrication and using it for gas separation. This copolymer is chosen for its relatively low cost. Based on the case of the polyamide, this copolymer should have improved characteristic compared to its individual homopolymer. This is noted by Marchese J. et al, (2003), “The ABS material has rubbery segments (butadiene) dispersed in a glassy matrix (styrene-co-acrylonitrile). This chemical structure suggests that ABS would allow obtaining relative high permeation fluxes (rubbery region) and relative high separation factors (glassy matrix)”

### **1.1 Objectives and scope of study**

The objective of this research is to:

- Fabricate ABS polymer membrane as dense and porous membrane in order to compare its performance to selected gases.

- Conduct test to measure its permeability using N<sub>2</sub>, CH<sub>4</sub> and O<sub>2</sub> to evaluate the performance of the membrane.

The scope of study includes:

Dense membrane for the experiment will be fabricated using solution-casting method. Porous membrane will be fabricated through phase inversion through solution precipitation. The solution will be precipitated by solvent evaporation. In order to achieve the porous structure, solvent mixture will be used to create a polymer rich and polymer poor phase that will ensure porous structure formation.

For measuring the membrane permeability, gas permeation test cell is used to measure the permeance of the gases. Based on the permeance of the gases used, ideal selectivity of the gases can be calculated and compared.

The next chapter (Chapter 2) will deal with theoretical background of separation process and membranes, focusing on the membrane type currently studied. Application of membranes and its transport mechanism is included.

Chapter 3 will outline the equipment and the methodology applied in membrane fabrication of the membrane studied, which is ABS copolymer membrane.

The following chapter, Chapter 4 will deal with results and discussion based on the experiment that had been conducted so far.

Chapter 5 will elaborate on the result and discussion.

The last chapter, Chapter 6 will go through the conclusion obtained based on the results and discussion.

## **CHAPTER 2**

### **LITERATURE REVIEW/THEORY**

#### **1.1 Separation process**

Separation processes are important to industries such as the pharmaceutical, petroleum, petrochemical, chemical, pulp, mineral, and other industries. It constitutes the majority capital cost and operating cost of those industries. Therefore it has major impacts on how profitable the industry is. Additionally to the obvious factors that influence the selection, design, and operation of separation processes are some items that seem unrelated. Its examples are an improved awareness of the impact of chemical processing on the environment, new products that need purity levels exceeding those typically encountered, the economics leading plant design, a recognition of the hazards associated with production of dangerous materials, and a rising consciousness of the consequence of political and natural boundaries on the availability of raw materials.

Driving force is important to a separation process. Since separation involves mass transfer, discussion regarding it is also important.

Common methods of separation in industry involve distillation, absorption and stripping, and extraction. However, membrane separation is gaining popularity. For a long time that membrane has been envisioned as a means to accomplish many types of separations. This goal has attracted several corporate entities to strive for the goal. Fresh developments in the membrane have brought these concepts to fruition, and the use of membrane-based separations is one of the most rapidly increasing interests in process technology.

#### **1.2 MEMBRANE**

##### **1.2.1 Major transport mechanism**

### 1.2.1.1 Gas separation membranes

Even though there are established technologies for separation, membrane technology is still being developed “due to the fact that membrane separation processes may offer more capital and energy efficiency compared to the conventional separation processes” (Lin and Chung, 2001). In addition, “advantages of membrane technology are its simplicity, i.e. no absorbent, which has to be regenerated; it can be easily retrofitted, modularized and scaled-up for several applications.” (Baker, 2000)

A membrane acts as a semi-permeable barrier. The CO<sub>2</sub> passes through this barrier more easily than other gases. In general, the rate at which a particular gas will move through the membrane can be determined by the size of the molecule, the concentration of gas, the pressure difference across the membrane and the affinity of the gas for the membrane material.

There are a number of mechanisms for gas separation in membranes:

1. Knudsen diffusion: gas components are separated based on the difference in the mean path of the gas molecules.
2. Molecular sieving: gas components are separated based on size exclusion, the size being the kinetic diameter of the gas molecules.
3. Solution-diffusion: the gases are separated by their solubility within the membrane and their diffusions through the dense membrane matrix. This is the usual separation mechanism for polymeric membranes (rubbers, polyimides, cellulose acetate).
4. Surface diffusion: gas molecules with higher polarity are selectively adsorbed onto the surface of the membrane and pass through the membrane by moving from one adsorption site to another...
5. Capillary condensation

The most common are molecular sieving and solution-diffusion.

The diversity of separation mechanism allows membrane to be developed with the best separation mechanism based on component to be separated.

#### 1.2.1.2 Gas Permeation Studies

“The permeation experiments always begin with nitrogen and ended with carbon dioxide. Feed side pressure was varied from 1 bar to 5 bars. The equipment set-up and was used to carry out the gas permeation measurement. The set-up consists of a feed gas tank, a pressure gauge of inlet gas, a dead-end membrane cell and a bubble soap flow meter. Membranes were located in the dead end membrane cell or module. This type of module allows the feed gas to flow into the membrane perpendicularly to the membrane position

Before performing the experiment, the gas permeation test unit was evacuated to less than 0.1 bars by vacuum pump for 1 hour to remove all residual gases remaining in the equipment. The feed gas was supplied directly from the gas tank, which is equipped with a pressure regulator. The feed gas pressure was set up within range of test pressure and the permeate stream was assumed to be at atmospheric pressure. In this permeation experiment, time (t) required to reach certain volume of gas in the permeate stream was observed and recorded. In addition, the volume of gas (V) in permeate stream was also measured using a bubble soap flow meter. The permeation of each gas through a membrane was measured twice at steady state condition”. (Iqbal, 2007)

Based on the volumetric measurements of the permeated gas, the volumetric flow rate,  $Q$ , was calculated as follows :

$$Q = \frac{V}{t}$$

This volumetric flow rate was then corrected to STP conditions (0°C and 1 atm) using the following equation

$$Q_{STP} = \frac{T_{STP}}{T} \times Q$$

in which  $T_{STP}$  and  $Q_{STP}$  referred to temperature (K) and volumetric of permeate gas ( $\text{cm}^3/\text{s}$ ) at STP condition. After conversion into STP condition, gas permeance,  $\frac{P}{l}$ , was then calculated using the following formula

$$\frac{P}{l} = \frac{Q_{stp}}{A \times \Delta p}$$

where  $\Delta p$  and  $A$  were trans-membrane pressure and effective membrane area, respectively. The  $\text{CO}_2/\text{CH}_4$  ideal selectivity (unitless),  $\alpha_{\text{CO}_2/\text{CH}_4}$ , of asymmetric membrane can be determined by dividing  $\text{CO}_2$  permeance,  $(P/l)_{\text{CO}_2}$ , over  $\text{CH}_4$  permeance,  $(P/l)_{\text{CH}_4}$ .

$$\alpha_{\text{CO}_2/\text{CH}_4} = \frac{(P/l)_{\text{CO}_2}}{(P/l)_{\text{CH}_4}}$$

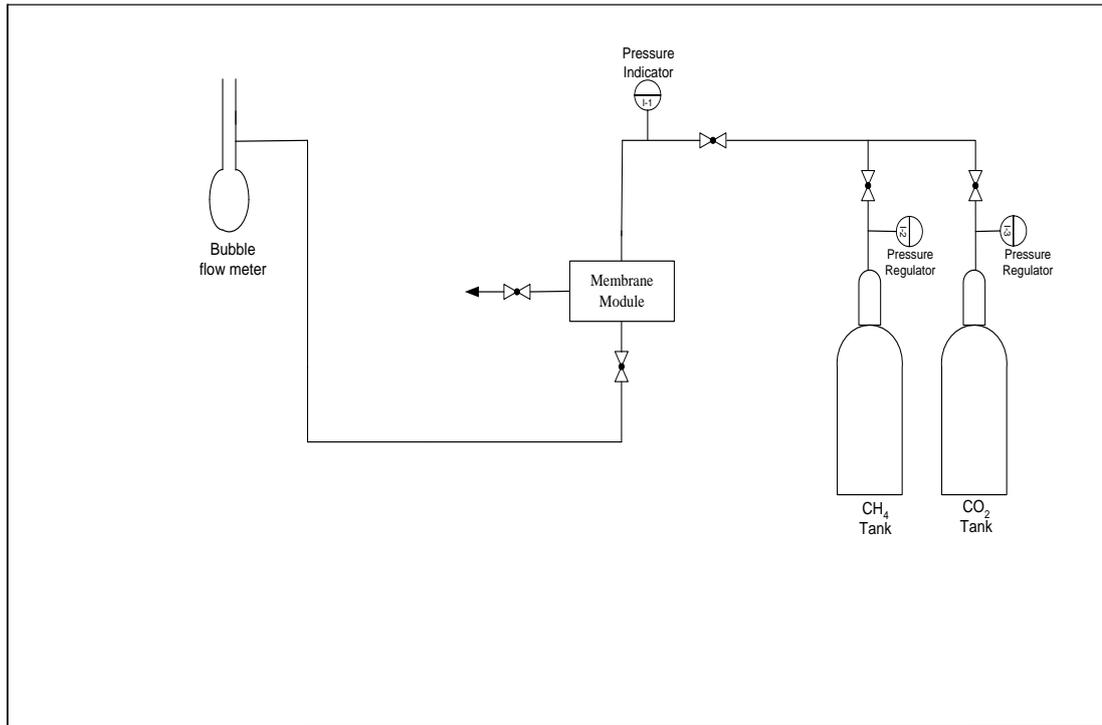


Figure 2-1: Schematic diagram for membrane permeation studies (Iqbal, 2007)

### 1.2.2 Mechanisms for membrane gas separation

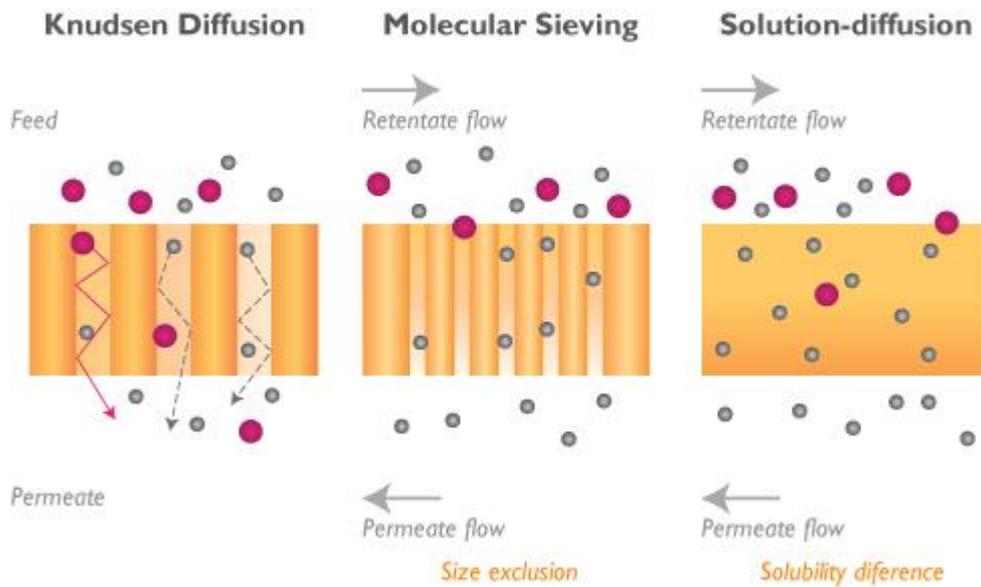


Figure 2-2: Schematic representation of three of the different possible mechanisms for membrane gas separation (CO2CRC, 2010)

### 1.2.3 Diffusion and Solubility coefficient

Transport through dense polymer may be considered as an activated process. Both diffusion and the solubility coefficient for the gas are temperature dependent, while

pressure is only observed for certain gases and material. For that, we can use Arrhenius equation to calculate diffusion and solubility coefficient.

$$D = D_o \exp(-E_d/RT) \qquad S = S_o \exp(-dH_s/RT)$$

$E_d$  = activation energy for diffusion

$dH_s$  = heat solution

$D_o$  and  $S_o$  is temperature independent constant

**Table 2-1: van't Hoff and Arrhenius parameters for the different penetrant/ABS systems (Marchese J. , Garis, Anson, Ochoa, & Pagliero, 2003)**

Gas	Activation energy for diffusion , $E_d$ (kcal/mol)
Oxygen	9.08
Nitrogen	10.24
Methane	10.31

**Table 2-2 : Penetrant physical properties (Marchese J. , Garis, Anson, Ochoa, & Pagliero, 2003)**

Gas	Kinetic diameter $\sigma_{kt}$ ( $\text{Å}$ )
Oxygen	3.46
Nitrogen	3.64
Methane	3.82

#### 1.2.4 Review of transport mechanism

The mechanism for gas transport through most of the ceramic membranes developed so far is Knudsen diffusion (Hyun et al, 1996). Therefore, its selectivity is not attractive enough for commercial application (Keizer et al , 1995).

For inorganic membrane with pore size smaller than 2 nm, there great potential for gas separation with reports that separation factor up to 60 (Keizer et al, 1995) at temperature of 100 °C. However, the fabrication process requires precise control and the result can vary drastically if any deviations exist in preparation process. Apart from that, surface modification can also be applied to alter the transport property of the membrane.

### **1.3 Type of membrane**

#### **1.3.1 Separation using inorganic membrane**

Keizer et al, (1995) pointed out that, according to manufacturer of inorganic membrane it has various advantages when compared to in organic membrane such as

- High pressure can be applied
- Possibilities of cleaning using steam
- Good back flushing possibilities to remove fouling

However, widespread use of inorganic membrane is hindered by lack of technology to form continuous and defect-free membranes, the extremely high cost for the membrane production, and handling issues (e.g., inherent brittleness)

#### **1.3.2 Separation using organic membrane**

Organic membrane is better than inorganic membranes in the terms of:

1. Desirable mechanical properties
2. Economical processing capabilities

The industry currently uses non-porous polymeric membrane based solution-diffusion mechanism for gas separation exclusively (Chung et al 2007).

## **1.4 Application of membrane**

Usually, carbon dioxide is removed from natural gas by using amine absorption system. However, the system has some drawback that leads to research into other technology. One of the alternatives already used is membrane separation. Membrane unit are usually smaller, and allow treatment directly at the wellhead. Treatment directly at the wellhead can reduce corrosion to the pipelines and eliminate risks with transporting to a centralized treatment plant. An example of these units is “Grace Membrane System” (Keizer et al, 1995)

## **1.5 Gas separation**

According to Keizer et al, (1995)

Important gas separation that currently employs membrane includes:

1. oxygen and nitrogen enrichment
2. hydrogen recovery
3. natural gas separation
4. the removal of volatile organic compounds from effluent streams

## **1.6 Gas separation using membrane**

### **1.6.1 Problems with using mixed matrix membrane**

“The adhesion between the polymer phase and the external surface of the particles appeared to be a major problem when glassy polymers are used in the preparation of such membranes. It seems that the weak polymer–filler interaction makes the filler tend to form voids in the interface between the polymer and the filler”. (Anson et al 2004). The voids reduce the selectivity of the membrane.

### **1.6.2 Surface modification using fluoro-silane coupling agent**

Fluorinated coupling reagents were preferred to alkyl ones because in each case CO<sub>2</sub> has a high solubility, but alkanes have “poor solubility in fluorinated compounds” (Abidi et al., 2006). Therefore, it is possible to increase the permeability of the inorganic membrane using fluorinated coupling reagents. There are similar research performed by Leger, et al, (1996), however, using different type coupling agent

(trichloro-octadecylamine). The advantage of this process is flexible and controllable, since it involves covalent bonding of organic compound to the surface of membrane.

## **CHAPTER 3**

### **METHODOLOGY**

#### **1.1 Organic membrane**

Organic membrane to be used will be fabricated in the lab. The membrane to be fabricated is ABS membrane. Two types of membranes will be fabricated which are porous and dense membrane.

#### **1.2 Gas used for permeability testing**

Since natural gas is mainly constituted of CH<sub>4</sub>, it is chosen to represent the natural gas to be tested. Another gas used is N<sub>2</sub> since it the gas to be separated the natural gas stream. O<sub>2</sub> is also used so that its permeability with N<sub>2</sub> can be compared. O<sub>2</sub> and N<sub>2</sub> represent the major constituents in air that is important for air enrichment.

#### **1.3 Dense Membrane preparation**

Membrane will be prepared using casting method. The method used derived from Marchese J. , et al, (2003). However, the details of the process is worked through trial and error process during membrane fabrication

Prior preparing the ABS solution, the ABS resin (Toyolac) is dried in the oven for 2 hours at 85 °C. The casting solution of ABS polymer is prepared by dissolving ABS resin in dichloromethane solvent (Cl<sub>2</sub>CH<sub>2</sub>) (Merck) according to 6 % concentration (w/v). For this fabrication purpose, 6.000 g of ABS is used. For the solvent, quantity used is 100 ml. During the addition process, the mixing speed is lowered to 30 rpm to prevent the ABS pallet from sticking to the bottom of the flask due to formation of vortex. The pallet is added slowly for the same reason. The solution is stirred continuously for at least 5 hours using magnetic stirrer at temperature of 25 °C at the highest speed possible while taking care preventing any ABS pallet from sticking to the bottom of flask and prevent the mixing process from happening. For this

experiment the mixture is stirred for 24 hour. During mixing, the round bottom flask is sealed using parafilm and aluminium foil to prevent the solvent from vaporizing during the mixing process. After the mixing time ended, all of the ABS resin had dissolved in the solvent, creating a cloudy mixture. Then, the solution was casted on a glass plate. Prior casting, masking tape is layered on the side of glass plate. The thickness of the casted membrane will be controlled by varying the thickness of masking tape used in order to contain the casting solution. About five layer of masking tape is used. After casting, excess casting solution is removed using stainless steel rod. The rod is rolled on the surface of the casting solution slowly and consistently. After removing the excess solution, the remaining solution is left to dry for about ten minutes before removing it from the membrane fabrication unit. The solution was left to dry at 25 °C for two days or until all traces of solvent removed.

#### **1.4 Porous membrane preparation**

The procedure for mixed membrane preparation was derived from Anson, et al, (2004) work. However, the details of the process is worked through trial and error process during membrane fabrication.

Prior preparing the ABS solution, the ABS resin (Toyolac) is dried in the oven for 2 hours at 85 °C. Solution of ABS polymer is prepared by dissolving ABS resin in dichloromethane solvent ( $\text{Cl}_2\text{CH}_2$ ) (Merck) and Tetrahydrofuran (Merck) with weight ratio 96:4 while 6 % concentration of ABS (w/w solvent). Total weight of solvent used is 100 g. For this fabrication purpose, 6.000 g of ABS is used. The solution is stirred continuously for at least 5 hours using magnetic stirrer at temperature of 25 °C. During the addition process, the mixing speed is lowered to 30 rpm to prevent the ABS pallet from sticking to the bottom of the flask due to formation of vortex. The pallet is added slowly for the same reason. For this experiment the mixture is stirred for 24 hour. During mixing, the round bottom flask is sealed using parafilm and aluminium foil to prevent the solvent from vaporizing during the mixing process. After the mixing time ended, all of the ABS resin had dissolved in the solvent, creating a cloudy mixture. Then, the solution was casted on a glass plate. Prior casting, masking tape is layered on the side of glass plate. The thickness of the casted membrane will be controlled by varying the thickness of masking tape used in order to contain the casting solution. About five layer of

masking tape is used. After casting, excess casting solution is removed using stainless steel rod. The rod is rolled on the surface of the casting solution slowly and consistently. After removing the excess solution, the remaining solution is left to dry for about ten minutes before removing it from the membrane fabrication unit. The solution was left to dry at 25 °C for two days or until all traces of solvent removed.

To remove the membrane fabricated for both the porous and the dense membrane, the masking tape is removed first. Then, using razor blade, the membrane is removed from the glass plate. When removing the membrane, utmost care is exercised in order to prevent the membrane from tearing. After the membrane is fully separated from the glass plate, it is further dried for a week.

## **1.5 Membrane characterization**

### **1.5.1 Permeability test**

Equipment that will be used for permeability test is gas permeability test unit. Based on Muhd Rusydi, (2009) the equipment setup is outlined. “The set-up consists of a feed gas tank, a pressure gauge of inlet gas, a dead-end membrane cell and a bubble soap flow meter. Membranes were located in the dead end membrane cell or module. This type of module allows the feed gas to flow into the membrane perpendicularly to the membrane position” (Muhd Rusydi, 2009).

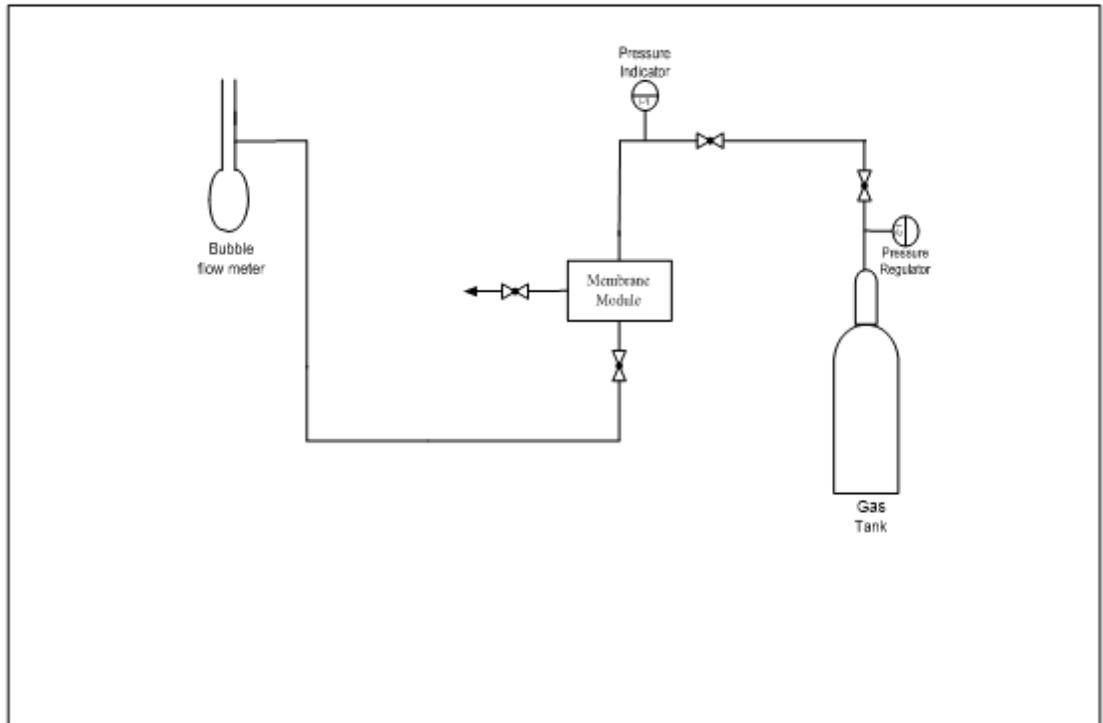


Figure 3-1: Gas permeability test setup

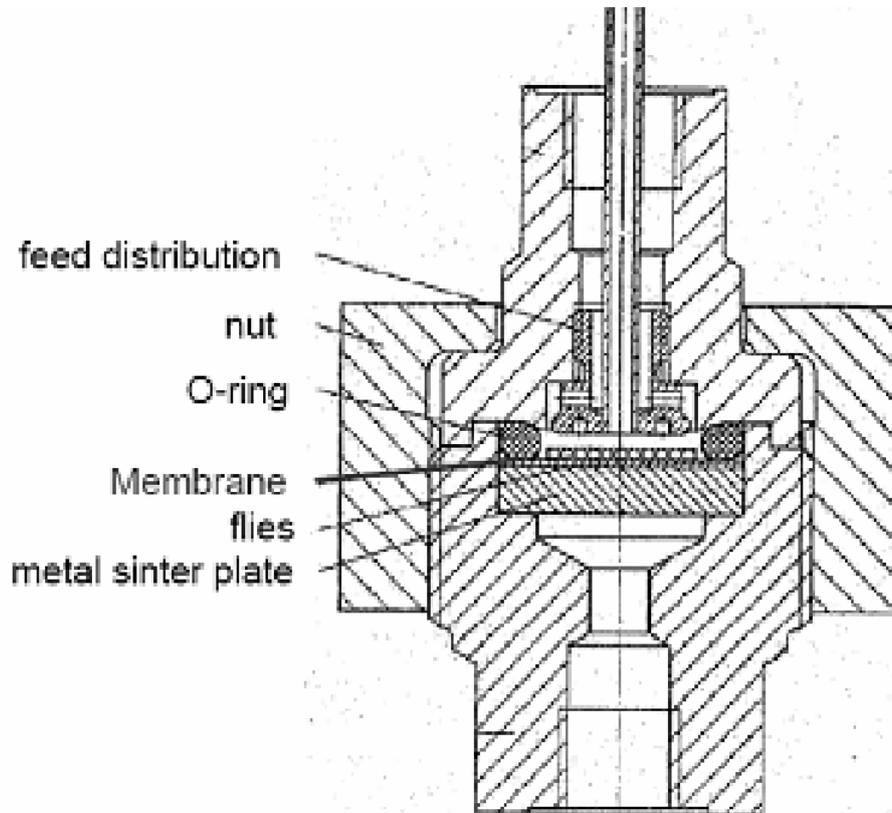


Figure 3-2: Detailed diagram for the membrane module (Budiyono, Kusworo, Ismail, Widiassa, Johari, & Sunarso, 2010)

Gas permeation measurement is conducted in Membrane Fabrication Laboratory, UTP. The permeation always begins with nitrogen and ended with methane. Feed side pressure is varied from one to three kg/cm<sup>2</sup>. The membrane is placed as shown in Figure 3-2.

Before performing the experiment, the gas permeation test unit was evacuated to less than one kg/cm<sup>2</sup> by vacuum pump for 1 hour to remove all residual gases remaining in the equipment. The feed gas was supplied directly from the gas tank, which is equipped with a pressure regulator. The feed gas pressure was set up within range of test pressure and the permeate stream was assumed to be at atmospheric pressure. In this permeation experiment, time (t) required to reach certain volume of gas in the permeate stream was observed and recorded. In addition, the volume of gas (V) in permeate stream was also measured using a bubble soap flow meter. The permeation of each gas through a membrane was measured twice once steady state condition is reached.

## **1.6 Tools required**

1. Beaker
2. Magnetic stirrer
3. Glass plate
4. Stainless steel roller
5. Weighing scale
6. Gas permeability unit
7. Gas Permeability Test Unit
8. Membrane fabrication unit

## **1.7 Chemicals**

1. General Purpose ABS resin Medium Impact(Toyolac)
2. Dichloromethane (Merck)
3. Tetrahydrofuran (Merck)
4. Purified Nitrogen (MOX)

5. Purified Oxygen (MOX)
6. Purified Methane (MOX)

### 1.8 Gantt Chart

No.	Detail/ Week	1	2	3	4	5	6	7	8	9	10	11	12	15	18	20
1	Project Work Continue	█	█	█												
2	Submission of Progress Report 1				●											
3	Project Work Continue				█	█	█	█								
4	Submission of Progress Report 2								●							
5	Seminar (compulsory)									█	█	█				
5	Project work continue							█	█	█	█					
6	Poster Exhibition												●			
7	Submission of Dissertation (soft bound)													●		
8	Oral Presentation														●	
9	Submission of Project Dissertation (Hard Bound)															●

● Milestone  
 █ Process

## CHAPTER 4

### RESULTS AND DICUSSION

#### 1.1 Permeability Test

Permeability test will be run for both of the porous and the dense membrane fabricated. Permeability test will be run for three gases which are Oxygen ( $O_2$ ), Nitrogen ( $N_2$ ), and Methane ( $CH_4$ ). The manipulated variables are type of gases and type of membrane and pressure. Pressures used are from one until three  $kg/cm^2$ .

#### 1.2 Volumetric Flow rate

##### 1.2.1 Dense membrane

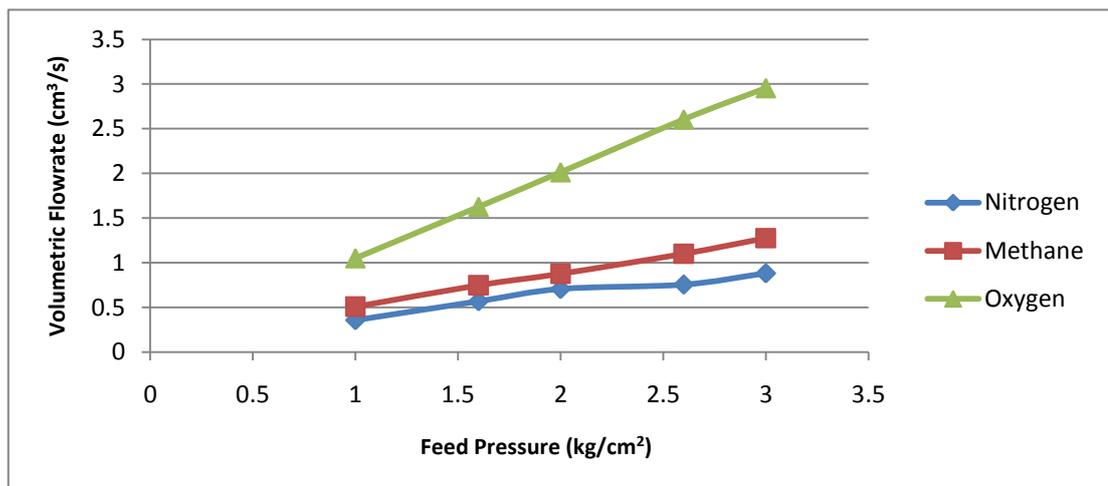


Figure 4-1: Volumetric Flowrate of gases vs Feed pressure

### 1.2.2 Porous Membrane

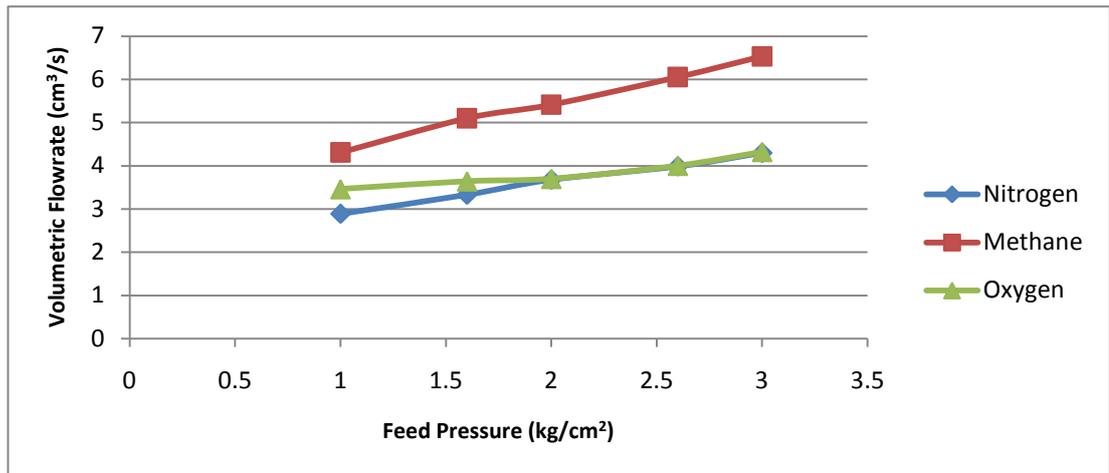


Figure 4-2: Volumetric Flowrate of gases vs Feed pressure

Comparing the volumetric flow rate for porous membrane and dense membrane, it can be noticed that dense membrane having higher gas flux compared to porous membrane for all three gases. Gas transport for dense membrane mostly occurs by solution diffusion while porous membrane having pores, gas transport will occur through the pores. Gas transport through the pores occurs faster than solution-diffusion. Since the main transport mechanism for porous membrane is through the pores, it has higher gas flux than dense membrane.

### 1.3 Permeance

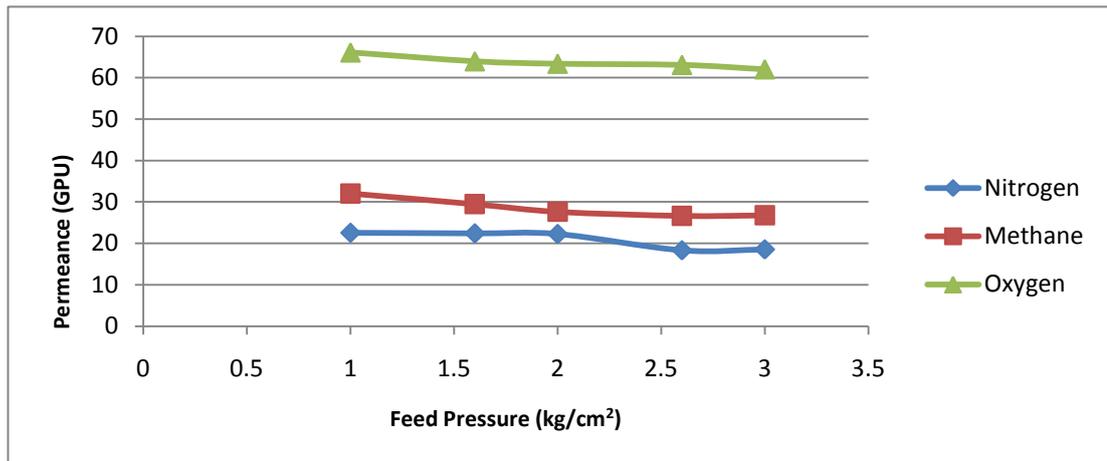


Figure 4-3: Gas Permeance vs Feed Pressure for Dense Membrane

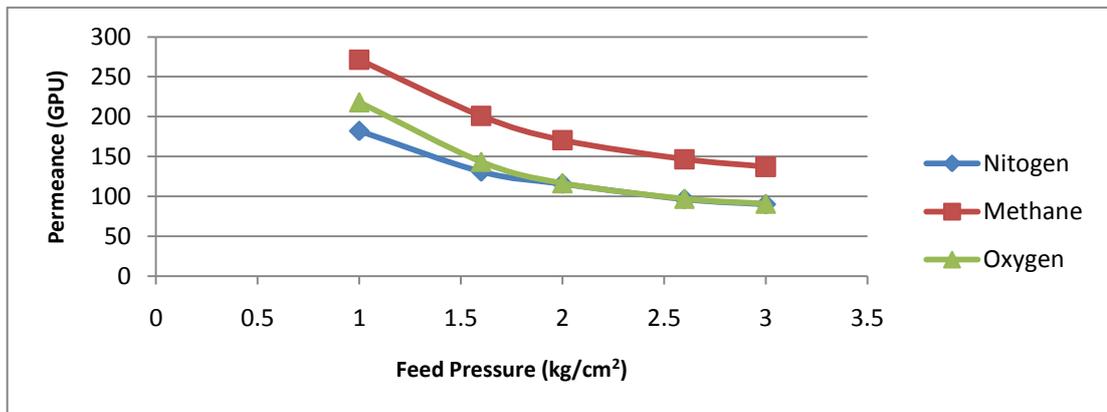


Figure 4-4: Gas Permeance vs Feed Pressure for Porous Membrane

For dense membrane,  $O_2$  have the highest permeability followed by  $CH_4$  and lastly  $N_2$ . This result is comparable to a study conducted by Marchese J., et al, (2003) where the ABS membrane of different manufacturer is used. Solution-Diffusion is a complex process that depends on the on the activation energy for diffusion ( $E_d$ ) the system. The permeability obtained have the same pattern as the  $E_d$  in Marchese J. , et al, (2003) where  $O_2$  have the lowest value of  $E_d$ , followed by  $N_2$  and  $CH_4$ . Based on the result, it is proven that the gas having lower value of  $E_d$  is easier to pass through the membrane through solution-diffusion as transport mechanism.

For porous membrane, the order of the permeance is  $CH_4 > O_2 > N_2$ . This pattern does not follow pattern of kinetic diameter as shown in Table 2-1. (Wang, et al,

1994) found similar pattern in permeance, and suggest that permeation was dominated by surface diffusion through grain boundary.

However, in Wang et al (1994), N<sub>2</sub> permeance is higher than O<sub>2</sub> permeance while in this study O<sub>2</sub> permeance is higher than N<sub>2</sub> permeance at low pressure. However, as pressure increase, permeance of N<sub>2</sub> increases at higher rate than O<sub>2</sub>. Based this observation, it can be conjectured that if the pressure is increased further, N<sub>2</sub> permeance will become higher than O<sub>2</sub> and therefore is similar with Wang, et al (1994) had found. The reason that N<sub>2</sub> permeance is higher than O<sub>2</sub> is because that at lower pressure, solution-diffusing is still the controlling transport mechanism for N<sub>2</sub> and O<sub>2</sub>. Only at higher pressure solution-diffusion is no longer the controlling transport mechanism.

Permeance of the gases decrease with increasing pressure for both membranes. As pressure increases, the gradient of adsorbed concentration decreases because the system is approaching the saturation capacity. Therefore, “the driving force for diffusion decreases, and the permeance declines” (Poshuta, et al, 1998). Based on the statement, all gases depend on adsorption process at a certain extent because all gases show decreasing pattern. In addition, the increase in pressure causes compression of pores in the membrane, reducing its permeability.

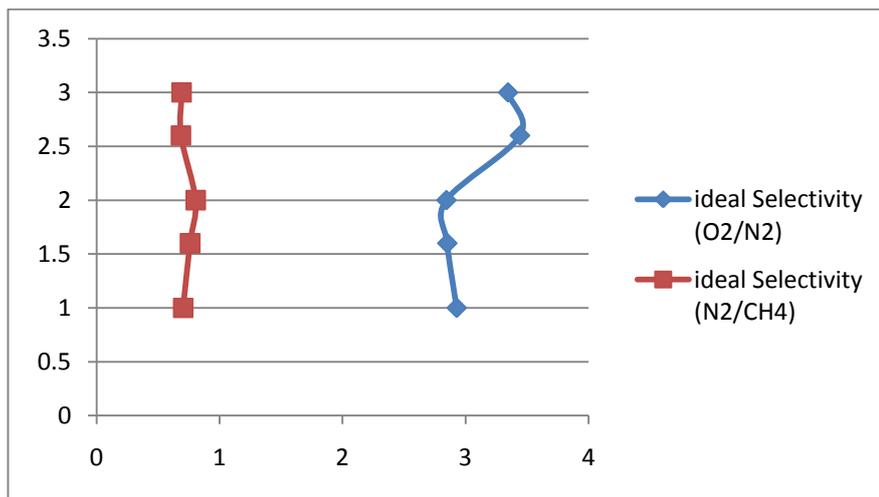
#### 1.4 Ideal Selectivity

Table 4-1 : Ideal Selectivity for porous membrane

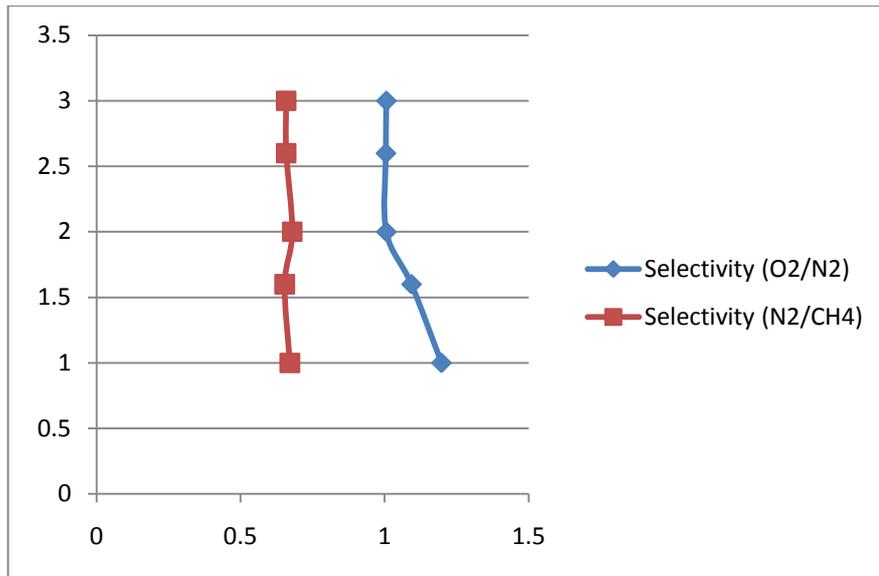
Pressure (kg/cm <sup>2</sup> )	Selectivity (O <sub>2</sub> /N <sub>2</sub> )	Selectivity (N <sub>2</sub> /CH <sub>4</sub> )
1	1.197300104	0.670714079
1.6	1.093226511	0.652898068
2	1.005919349	0.679293858
2.6	1.004403523	0.658429653
3	1.006052745	0.657928664

**Table 4-2 : Ideal Selectivity for dense membrane**

Pressure (kg/cm <sup>2</sup> )	Selectivity (O <sub>2</sub> /N <sub>2</sub> )	Selectivity (N <sub>2</sub> /CH <sub>4</sub> )
1	2.928121721	0.704354058
1.6	2.851340374	0.760968661
2	2.844064386	0.806154935
2.6	3.442708333	0.687216339
3	3.345642541	0.692273731



**Figure 4-5 : Ideal Selectivity for dense membrane**



**Figure 4-6 : Ideal Selectivity for porous membrane**

The selectivity of O<sub>2</sub>/N<sub>2</sub> and N<sub>2</sub>/CH<sub>4</sub> are compared because of its importance in industry. The selectivity for porous membrane is lower than dense membrane. The reason for this is transport mechanism in dense membrane (solution-diffusion) have better selectivity compared to transport mechanism in porous membrane.

For dense membrane, the highest selectivity is at 2.6 kg/cm<sup>2</sup> for O<sub>2</sub> /N<sub>2</sub> and the highest selectivity for N<sub>2</sub>/CH<sub>4</sub> are at pressure 2 kg/cm<sup>3</sup>.

For porous membrane, the highest selectivity is at pressure of 1 kg/cm<sup>2</sup> for O<sub>2</sub>/N<sub>2</sub> and for N<sub>2</sub>/CH<sub>4</sub>, the highest selectivity is at 2 kg/cm<sup>2</sup>.

## **CHAPTER 5**

### **CONCLUSION AND RECOMMENDATION**

#### **1.1 Conclusion**

From ABS, dense and porous membrane had been prepared. For dense membrane, solution casting method was used while for porous membrane, dry phase inversion in used.

Dense membrane has lower overall flux compared to porous membrane since dense membrane could only rely on solution-diffusion for gas transport while porous membrane have pores that allow more gas to pass through the membrane.

Ideal selectivity of dense membrane is higher than ideal selectivity of dense membrane since dense membrane rely on solution-diffusion for as main gas transport mechanism while porous membrane depends on pores. Solution-diffusion has better selectivity since it depends on chemical properties instead of physical properties which most porous membrane transport mechanism rely on.

#### **1.2 Recommendations**

For future works, several recommendations is outlined below:

##### **1. Conduct permeability experiment using mixed gas**

By using mixed gas during permeability experiment, real selectivity of the gases can be established.

##### **2. Use ABS pallet with higher strength**

The ABS pallet currently used cannot withstand high pressure at feed stream. By using ABS pallet with higher strength, higher pressure for feed stream can be used.

## CHAPTER 6

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Marchese, J., Garis, E., Anson, M., Ochoa, N. A., & Pagliero, C. (2003). Gas sorption, permeation and separation of ABS copolymer membrane. *Journal of Membrane Science* , 185-197.

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**CHAPTER 7**  
**APPENDIX 1**

Raw Data

Porous Membrane

Nitrogen

30

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cm <sup>-3</sup> .s <sup>-1</sup> .cmHg <sup>-1</sup> )
1	9.8067E+04	73.55592	50	17.26	17.33	0.0509	2.8910 09	2.6308181 56	182.0501247
1.6	1.5691E+05	117.689472	50	14.98	15.04	0.0509	3.3311 13	3.0313124 58	131.1024695
2	1.9613E+05	147.11184	50	13.66	13.53	0.0509	3.6778 23	3.3468186 83	115.7983416
2.6	2.5497E+05	191.245392	50	12.54	12.55	0.0509	3.9856 52	3.6269430 05	96.53116189
3	2.9420E+05	220.66776	50	11.63	11.64	0.0509	4.2973 79	3.9106145 25	90.20360715

Methane

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cm <sup>-3</sup> .s <sup>-1</sup> . cmHg <sup>-1</sup> )
1	9.8067E+04	73.55592	50	11.64	11.56	0.0509	4.3103 45	3.9224137 93	271.4273196
1.6	1.5691E+05	117.689472	50	9.81	9.79	0.0509	5.1020 41	4.6428571 43	200.8008232
2	1.9613E+05	147.11184	50	9.17	9.3	0.0509	5.4141 85	4.9269085	170.468701
2.6	2.5497E+05	191.245392	50	8.38	8.14	0.0509	6.0532 69	5.5084745 76	146.6081629
3	2.9420E+05	220.66776	50	7.64	7.67	0.0509	6.5316 79	5.9438275 64	137.1024127

31

Oxygen

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cm <sup>-3</sup> .s <sup>-1</sup> . cmHg <sup>-1</sup> )
1	9.8067E+04	73.55592	50	14.59	14.3	0.0509	3.4614 05	3.1498788 51	217.9686333
1.6	1.5691E+05	117.689472	50	13.76	13.7	0.0509	3.6416 61	3.3139111 43	143.3246954
2	1.9613E+05	147.11184	50	13.53	13.5	0.0509	3.6995 93	3.3666296 71	116.4837924
2.6	2.5497E+05	191.245392	50	12.58	12.4	0.0509	4.0032 03	3.6429143 31	96.95623907
3	2.9420E+05	220.66776	50	11.47	11.66	0.0509	4.3233 9	3.9342844 79	90.74958661

Dense membrane

Nitrogen

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cm <sup>-3</sup> .s <sup>-1</sup> .cmHg <sup>-1</sup> )
1	9.8067E+04	73.55592	10	27.97	27.84	0.0509	0.3583 59	0.326106	22.56626
1.6	1.5691E+05	117.689472	10	17.5	17.6	0.0509	0.5698 01	0.518519	22.42562
2	1.9613E+05	147.11184	10	14.07	14.2	0.0509	0.7074 64	0.643792	22.2749
2.6	2.5497E+05	191.245392	10	13.1	13.34	0.0509	0.7564 3	0.688351	18.32048
3	2.9420E+05	220.66776	10	11	11.65	0.0509	0.8830 02	0.803532	18.53455

Methane

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cmHg <sup>-1</sup> .s <sup>-1</sup> )
1	9.8067E+04	73.55592	10	19.47	19.84	0.0509	0.5087 76	0.462987	32.03823
1.6	1.5691E+05	117.689472	10	13.21	13.5	0.0509	0.7487 83	0.681393	29.46983
2	1.9613E+05	147.11184	10	11.43	11.36	0.0509	0.8775 78	0.798596	27.63104
2.6	2.5497E+05	191.245392	10	9	9.17	0.0509	1.1007 15	1.001651	26.65896
3	2.9420E+05	220.66776	10	7.74	7.94	0.0509	1.2755 1	1.160714	26.77344

Oxygen

pressure(kg/c m <sup>2</sup> )	pressure (pascal)	Pressure (cmHg)	Volume (cm <sup>3</sup> )	Time (s) 1st run	Time (s) 2nd run	area <sup>-1</sup> (cm <sup>-3</sup> )	Q (cm <sup>3</sup> )	Q(stp) (cm <sup>3</sup> )	GPU (cm <sup>3</sup> .cmHg <sup>-1</sup> .s <sup>-1</sup> )
1	9.8067E+04	73.55592	10	9.79	9.27	0.0509	1.0493 18	0.954879	66.07675
1.6	1.5691E+05	117.689472	10	6.24	6.07	0.0509	1.6246 95	1.478473	63.94307
2	1.9613E+05	147.11184	10	4.91	5.03	0.0509	2.0120 72	1.830986	63.35125
2.6	2.5497E+05	191.245392	10	3.74	3.94	0.0509	2.6041 67	2.369792	63.07205
3	2.9420E+05	220.66776	10	3.4	3.37	0.0509	2.9542	2.688331	62.00998

**CHAPTER 8**  
**APPENDIX 2**

Membrane fabrication



Solution  
mixing



Casting



Final Drying



Drying