EFFECT OF FEED TEMPERATURE AND COMPOSITION OF PROPANE ON THE SEPARATION OF CARBON DIOXIDE – METHANE – PROPANE BY USING HOLLOW FIBER MEMBRANE

By

RAFIQ HAKIM BIN RAZALI 12830

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

JAN 2013

Universiti Teknologi PETRONAS Bandar Seri Iskandar 31750 Tronoh Perak Darul Ridzuan

CERTIFICATION OF APPROVAL

EFFECT OF FEED TEMPERATURE AND COMPOSITION OF PROPANE ON THE SEPARATION OF CARBON DIOXIDE – METHANE – PROPANE BY USING HOLLOW FIBER MEMBRANE

by

Rafiq Hakim Bin Razali

A project dissertation submitted to the

Chemical Engineering Program

Universiti Teknologi PETRONAS

in partial fulfillment of the requirement for the

Bachelor of Engineering (Hons)

(Chemical Engineering)

Approved by,

(DR LAU KOK KEONG)

Date: 15th September 2013

UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

JAN 2013

CERTIFICATION OF ORIGINALITY

This is to certify that I, Rafiq Hakim Bin Razali (I/C No: 900622-01-6351), 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.

RAFIQ HAKIM BIN RAZALI

- Student ID : 12830
- I/C No : 900622-01-6351
- Date : 15th September 2013

ABSTRACT

Carbon dioxide, CO_2 is known with its corrosive properties when react with water. Therefore, CO_2 is needed to be removed from the natural gas to avoid it from damaging the pipelines and equipment. Nowadays, there are a lot of method to separate CO_2 from natural gas such as physical absorption, chemical absorption, adsorption process and also membrane technology. A simple technology such as membrane technology is suggested to be applied to remove CO_2 from natural gas especially at remote and unattended situation like offshore area. This is because separation using membrane technology is known with its high efficiency and less cost. However, the research conducted on multi-component separation of CO₂ from natural gas using hollow fiber membrane is very limited nowadays. Most of them are more focusing on binary component separation. So, this project is conducted to study evaluate the permeability of multi-component separation of CO₂-Methane-Propane by changing the parameters which are feed temperature and composition of propane. First of all, the hollow fiber membrane is potted into the module shell before tested at the test rig. The experiment is conducted by using Carbon Dioxide Separation Membrane Unit (CO2SMU) facilities. The result obtained from this experiment will be used to analyze between the membrane permeability and the parameters. Based on result, the permeability is decrease as the feed temperature increase. Meanwhile, as the propane composition is increases, the permeability is also decrease.

ACKNOWLEDGEMENT

Thanks to God, whom with His willing giving me the opportunity to complete this Final Year Project. First and foremost, I would like to express my deepest gratitude to my helpful supervisor Dr. Lau Kok Keong, who has guided and support me during these two semester sessions to complete this project.

I would also want to thank all lecturers and staffs of Chemical Engineering Department for their co-operations, suggestions and time responding to my inquiries along the way. Deepest thanks and appreciation to my beloved parents.

Not to forget, to all my friends and work mates especially Tan Et Kuan and Ponmani Sivagnanam for their cooperation, encouragement, constructive suggestion and full of support for this project completion, from the beginning till the end. Thanks to everyone who has been contributing by supporting my work during the final year project progress till it is fully completed.

TABLE OF CONTENT

CERTIF	FICATION OF APPROVAL	ii
CERTIF	FICATION OF ORIGINALITY	iii
TABLE	E OF CONTENT	vi
CHAPT	ГЕR 1	9
INTRO	DUCTION	9
1.1	BACKGROUND OF STUDY	9
1.2	PROBLEM STATEMENT	10
1.2.	2.2 Significant of the project	10
1.3	OBJECTIVE	11
1.4	SCOPE OF STUDY	11
1.5	THE RELEVANCY OF PROJECT	11
1.6	FEASIBILTY OF THE PROJECT	12
CHAPT	ГЕR 2	13
LITERA	ATURE REVIEW	13
2.1 Lit	terature Review	13
2.1.	.1 Membrane Definition and Classifications	13
2.1.	2 Hollow Fiber Membrane	14
2.1.	3 Past researches done on CO_2 separation using membrane	15
2.1.	.4 Research Gap	21
2.2	Theory	22
2.2.	P.1 Flux and Permeability Concept	22
2.2.	2.2 Transport Mechanism	22
CHAPT	ГЕR 3:	25
METHO	ODOLOGY	25
3.1	Research Methodology and Project Activities	25
3.2	Experimental Procedures/Approach	25
3.2.	Lab scale hollow fiber membrane module	26
3.2.	Potting fibers in the module	27
3.2.	9.3 Gas separation testing unit	29

3.3 The effect of feed temperature	30
3.4 The Effect of Propane composition	31
3.5 Gantt Chart	32
3.5.1 Final Year Project 1	32
3.5.2 Final Year Project 2	33
CHAPTER 4:	36
RESULT AND DISCUSSION	36
4.1 Effect of Feed Temperature	36
4.3 Effect of Propane Composition	38
CHAPTER 5:	40
CONCLUSION	40
REFERENCES	41
APPENDIX	43

LIST OF FIGURES

Figure 1: SEM image on hollow fiber membrane. a: cross-sectional; b: inner surface; couter surface. (Kirk-Othmer, 2004)
Figure 3: Schematic diagram of hollow fiber membrane separation (Kirk-Othmer, 2004) 14
Figure 4: Surface Diffusion (Basile, 2011)
Figure 5: Capillary Condensation (Basile, 2011)
Figure 6: Molecular Sieving (Basile, 2011)
Figure 7: Multi-layer Diffusion (Basile, 2011)
Figure 8: Poiseuille (Viscous) Mechanism (Basile, 2011)
Figure 9: Knudsen Mechanism (Basile, 2011)
Figure 12 : The schematic diagram depicting the general approach in this project
Figure 13: The hollow fiber membrane module structure
Figure 14: The lab-scale hollow fiber membrane module structure
Figure 15: Budle preparation and module assembly
Figure 16: Epoxy adhesive
Figure 17: Removing solidified epoxy resin using hand drill
Figure 18: Flow sheet of gas separation testing unit for experimental validation
Figure 19: Graph of Permeance vs Feed Temperature
Figure 23: Graph of Gas Permeance vs Propane Composition

LIST OF TABLES

Table 1: Effect of Feed Temperature on Gas Permeance	. 45
-	
Table 2: Effect of Propane Composition on Gas Permeance	. 48

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND OF STUDY

As the year increase, the demand for natural gas increases rapidly. Natural gas consist of methane, ethane, propane, butane, higher molecular weight hydrocarbon and other impurities. Carbon dioxide, CO₂ is acid gas contains in natural gas. High CO₂ content in natural gas is one of the major problem that we are facing nowadays. CO₂ will become very corrosive when react with water and it can destroy pipelines and equipment.

Separation of carbon dioxide, CO_2 can be done in many of ways. Numerous of CO_2 separation processes have been developed and improved from time to time in order to optimize the capital cost and operating cost, to meet gas specifications, and for environmental purpose. Among the processes available for CO_2 separation are absorption processes (chemical and physical absorption), adsorption process (solid surface), hybrid solution (mixed physical and chemical solvent) and also physical separation (cryogenic separation and membrane).

However, to separate CO_2 at offshore situation need a more simple separation technology. In this case membrane technology is suitable to be applied. Gas separation using membrane technology is an essential unit operation in various industrial separations including petrochemical, gas processing and also biotechnology. Nowadays, hollow fiber membrane module is becoming widely used in the industrial separations because of its low capital cost, less space requirement, high efficiency, lower energy requirement, and chemical-free operation. Membrane can be defined as a thin barrier placed between two mediums or phases which allow one or more constituents to selectively pass from one medium to another while retaining the rest. (Dutta, 2007).

1.2 PROBLEM STATEMENT

1.2.1 Problem identification

 CO_2 need to be removed from natural gas before the gas is transported through the pipelines. It can destroy pipelines and equipment because the mixture of CO_2 and water is very corrosive. A simple process technology to remove CO_2 from natural gas is needed to be applied at remote, unattended, or offshore situations. In this scenario, hollow fiber membrane seems the best method to separated CO_2 from the natural gas. Many researches have been done to study the separation CO_2 from natural gas. However, most of them are focusing on binary component separation. So, this research is done to focus on multi component separation (CO_2 -Methane-Propane) by using hollow fiber membrane.

1.2.2 Significant of the project

The aim of the project is to study the relationship between the changes in the in parameters (feed temperature and propane concentration) and the efficiency of the CO_2 removal using a multi-component hydrocarbon feed; CO_2 , CH_3 and C_3H_8 . The experiment will be conducted using feed of different propane concentration and feed temperature to obtain data for the study. This can be used to evaluate and optimize the membrane system used to remove CO_2 .

1.3 OBJECTIVE

- 1. To evaluate the permeability of CO₂-Methane-Propane separation at different feed temperature
- 2. To evaluate the permeability of CO₂-Methane-Propane separation at different Propane composition

1.4 SCOPE OF STUDY

The scope of this project is to conduct a literature review on removal of CO_2 from natural gas (methane and propane) stream using hollow membrane and the effect of feed temperature and propane composition. The next step is to proceed with conducting experiment on fabrication of hollow fibre membrane for different parameters that affect the separation process. Through this project student is exposed to explore research problems and build research objectives, applying appropriate methodology, analysing and interpreting data obtained from the experiment, troubleshooting any predicaments occur ad also reporting the findings.

1.5 THE RELEVANCY OF PROJECT

Availability of a simple process technology that can be applied in remote, unattended, or offshore situations is highly desirable. Membrane based system provide a simple, low-cost, compact solution to separate CO_2 from natural gas. This project is conducted so that the performance of hollow fiber membrane can be investigated based on the parameter which are the feed temperature and propane composition in the mixture of CO_2 -Methane-Propane.

1.6 FEASIBILTY OF THE PROJECT

This project encompasses experimental work. The experimental work will be conducted by using Carbon Dioxide Separation Membrane Unit (CO2SMU) which is available at Block N in Universiti Teknologi Petronas. This project can be done within 8 months given that everything goes well. The objective can be achieved if the procedures are accurately followed.

CHAPTER 2

LITERATURE REVIEW

2.1 Literature Review

There were a lot of literatures found about the study on carbon dioxide separation from natural gas stream by using hollow fiber membrane technology. Therefore, these literatures would be very helpful reference and guideline to study on how the feed temperature and propane composition affect the CO_2 – methane – propane separation by using hollow fiber membrane.

2.1.1 Membrane Definition and Classifications

Membrane is defined as selective barrier between two phases that has ability to transport one component than the other (Mulder, 1996). Membrane applications attracting a broad range of applications such as for sea water desalination, wastewater treatment, and ultrapure water production for semiconductor industry and nitrogen enrichment from air. Each of these applications requires specific type of membrane morphology to ensure the effective separation (Iqbal, 2007).

Generally, membrane morphologies are classified into two categories, symmetric and asymmetric membrane (Mulder, 1996). Symmetric membrane is the membranes that have essentially same structure and transport properties throughout its thickness while asymmetric membrane is a membrane constituted of two or more structural planes of non-identical morphologies (Koros, 1996).

2.1.2 Hollow Fiber Membrane

Hollow fiber membrane can be defines as a capillary having an inside diameter of larger than 25 μ m and outside diameter less than 1mm; and its wall functions as semipermeable membrane (Kirk-Othmer, 2004). Commonly, hollow fiber membranes are used as cylindrical membranes that permit selective exchange of materials across the walls. However they can also be used as reactors to chemically modify a permeate as it diffuses through a chemically activated hollow fiber wall.



Figure 1: SEM image on hollow fiber membrane. a: cross-sectional; b: inner surface; couter surface. (Kirk-Othmer, 2004)





Nowadays, the applications of hollow fiber membrane in industrial separation become wider due to its capabilities such as high efficiency, lower energy requirement, chemical-free operation, etc. (Katoh, 2011). Conventional hollow fiber membrane models in process simulators usually assume constant membrane permeance (independent temperature and pressure).

Recently, Faizan et al (2013) proposed a new simulation model to study the effect of temperature and pressure of the membrane permeates. Simulations are performed to study the effect of stage cut and temperature drop on membrane permeance of CO_2 and CH_4 . As a result, it can be seen that permeance of both CO_2 and CH_4 decreases with the increase in stage cut. This is because of the increment of stage cut will increase the temperature drop which results in change of CO_2 and CH_4 permeance being dependent on temperature. Feed pressure also affects the membrane permeance. This is due to the mass transport of the membranes for the separation of CO_2 and CH_4 mixtures is determined by competitive sorption and plasticization.

No	Author	Year	Objectives		Findings
1	Qin et al.	2005	To investigate the effect of shear rate within the spinneret on CO ₂ /CH ₄ separation performance.	1.	The CO ₂ /CH ₄ selectivity of the copolyimide dense film decreased significantly with an increase in temperature. The performance of as-spun fibers was obviously influenced by the shear rate during spinning. For uncoated fibers, permeances of CH ₄ and CO ₂ decreased with increasing shear rate, while selectivity of CO ₂ /CH ₄ sharply increased with shear rate until the shear rate reached 2169 s–1 and then

2.1.3 Past researches done	on CO ₂ separatio	n using membrane
----------------------------	------------------------------	------------------

					the selectivity leveled off.
				3.	After silicone rubber
				5.	
					coating, permeances of CH_4
					and CO_2 decreased, the
					selectivity of CO ₂ /CH ₄ was
					recovered to the inherent
					selectivity of its dense film.
					Both the permeances and
					selectivity with increasing
					shear rate followed their
					same trends as that before
					the coating.
				4.	There was an optimal shear
					rate at which a defect-free
					fiber with a selectivity of
					CO ₂ /CH ₄ at 42.9 and
					permeance of CO ₂ at 53.3
					GPU could be obtained after
					the coating.
2	Chenar et al.	2006	To investigate the	1.	The presence of water vapor
			effect of water vapor		in the feed stream
			on the performance		significantly reduced the
			of commercial polyphenylene oxide		permeation rates of both CO_2 and CH_4 through
			and Cardo-type		hydrophilic Cardo-type
			polyimide hollow		polyimide hollow fiber
			fiber membranes in		membrane. The separation
			CO ₂ /CH ₄ separation		factor and selectivity,
			applications		however, remained the same because the permeation rates
					of both gases were declined
					to the same extent.
				2.	A small decline in the
					permeation rate of CO ₂ was
					observed through PPO hallow fiber membrane, but
					CH ₄ permeation rate was not
1				1	affected significantly. As a

					result, a decline in the separation factor and selectivity of this membrane was observed.
3	Mohd Jefri b Yusof Atchariyawut et al.	2010	1.To study on carbon dioxide separation from natural gas using inorganic membrane.2.To study on characteristics of the inorganic membrane when treated with natural gas on certain operating conditions.3.To study on permeability and ideal selectivity of carbon dioxide on inorganic.To investigate the effect of gas and liquid velocity, 	1. 2. 3. 4. 1. 2.	selectivity than methane, average value of 3.5 which mean that carbon dioxide permeate through membrane 3.5 faster than methane.
5	Nurul Safiah Binti Mat Dagang	2010	1. To synthesize membrane with higher porosity	1.	Porosity percentage increases when starch drop is increased.

	1					
				than 22.53 % (up	2.	It shows that starch can be
				to 25-30 %).		used as a binder to control
			2.	To synthesize		the porosity of alumina
				membrane that		support.
				provides high	3.	Flux is also increases when
				permeability and		the inlet flow rate and inlet
				selectivity higher		pressure are increased.
				than 1.37 (up to		However, flux decreases as
				1.66 ideal		the dip coating hours is
				separation		increased.
				factor).	4.	The permeability decreases
			3.	To evaluate the		as the inlet pressure is
				performance of		increased due to
				the synthesized		concentration of gas
				membrane.		molecules approaches
						saturation in the inlet side.
6	Shuji	2007	То	study the	1.	In mixed-gas permeation
	Himeno		syı	nthesis and		experiments using the sweep
			pe	rmeation		method, the DDR-type
			pro	operties of DDR-		zeolite membrane showed
			Ту	pe Zeolite		high selectivity for
			M	embrane for		CO ₂ /CH ₄ mixtures of 200
			CC	D ₂ /CH ₄ separation		and high CO ₂ permeance.
			ga	seous mixture.	2.	DDR zeolite membranes
						were compared to
						other zeolite membranes for
						evaluation of their
						CO ₂ /CH ₄ selectivity and
						CO ₂ permeance; the DDR-
						type zeolite membranes
						show better CO ₂ /CH ₄
						separation and
1						~ ~
						CO ₂ permeance.

7	Hedayat et	2011	To investigate the	1.	The presence of CO ₂ in feed
	al.		effect of temperature,		gas decreased the H2s
			pressure, gas and		removal. However, increase
			liquid flow rates,		of CO ₂ concentration had a
			absorbent		stronger negative influence
			concentration and		on removal efficiency of
			acid gas content of		itself compared with H ₂ S.
			the feed.	2.	Pressure had a positive
					effect on the removal
					efficiency.
				3.	Temperature had no
					significant effect on H ₂ S
					removal, but decreased the
					CO ₂ removal and thus
					enhanced H ₂ S selectivity.
8	Schrier	2012	To study carbon dioxide separation with a two- dimensional polymer	1. 2. 3. 4.	Developed a Langmuir- adsorption model to calculate the effect of surface adsorption of gases on membrane permeance Simulated the CO2 separation from N ₂ and CH ₄ using hydrocarbon polymer, PG-ES The CO ₂ permeance is 3 \times 10 ⁵ gas permeation units (GPU) The CO ₂ /N ₂ selectivity is 60, and the CO ₂ /CH ₄ selectivity exceeds 500
9	Jahn et al.	2013	To study CO ₂ flux obtained from different membrane processes through experiments	1.	$\begin{array}{c cccc} Made & comparison & of \\ CO_2 & flux & obtained & from \\ different & membrane \\ processes & through \\ experiments & using & pure & gas \\ and & also & binary & gas & of & CO_2- \\ H_2 & and & CO_2-CH_4 & \\ CO_2 & flux & across \\ selective & membrane & is \\ higher & for & inorganic \\ membrane & and & membrane \\ contactors & compared & to \\ \end{array}$

				 polymer membrane 3. CO₂ flux is decreasing with increasing permeate side pressure whereas the CO₂ flux increases with increase in feed pressure
10	Yang et al.	2009	the effect cross flow model and co-current flow model	 3.1 Difference between the cross-flow model and the other co-current model is insignificant 3.2 Increasing feed side pressure and decreasing permeate side pressure, decreases the membrane area required and increases CH₄ recovery 3.3 • Achieved CH₄ recovery 3.3 • Achieved CH₄ recovery of more than 98% and product purity of more than 98% by the single-stage system using membrane selectivity of 20%
11	Scholes et al.	2012	To study the current and future potential of polymeric membranes in acidic gas removal, heavy hydrocarbon recovery, water dehydration as well as nitrogen and helium separation	 Polymeric membrane in natural gas separation have been successfully proven and efficient processes have been implemented Membrane gas separation will grow in this market as the technology is increasing seen as commercially competitive and less high risk

2.1.4 Research Gap

From the literature review, it can be concluded that membrane system is one of the best methods to separate CO_2 from natural gas due to its advantages compared other technology in terms of operational cost and space requirement. Numerous of studies have been done on membrane separation technology.

The research are basically divided into simulation of development of mathemathical model, membrane performance, development of new membrane and optimization of existing membrane.

However, there are still lack of researches done on multi component separation. Most of them are focused on binary component separation. Therefore, there is still a large gap between the lab-scale and the real application in the industry. There is uncertainty over the effectiveness of these membranes under industrial conditions where the feed gas is made up of multi component. It can be clearly seen that there is a gap in understanding the effect of multi component feed on the membrane performances.

2.2 Theory

2.2.1 Flux and Permeability Concept

According to Munirah (2012), flux is one of the membrane transport where flux is expressed by the unit of [mol.m⁻².s⁻¹Pa⁻¹]. As opposed to flux which is normalized per unit pressure, permeation or permeability is normalized per unit thickness [mol.m.m⁻².s⁻¹Pa⁻¹].

2.2.2 Transport Mechanism

Basile (2011) stated that there are a few transport mechanisms for membrane such as surface diffusion, Poiseuille (Viscous) mechanism, Knudsen mechanism et cetera.

Surface Diffusion



Figure 3: Surface Diffusion (Basile, 2011)

This mechanism is achieved when one of the permeating molecules is adsorbed on the pore wall. This type of mechanism can reduce the effective pore dimensions obstructing the transfer of different molecular species. Capillary Condensation



Figure 4: Capillary Condensation (Basile, 2011)

This type of mechanism takes place when one of the components condenses within the pores due to capillary forces. Generally, the capillary condensation favours the transfer of relatively large molecules.

Molecular Sieving



Figure 5: Molecular Sieving (Basile, 2011)

This takes place when pore diameters are very small, allowing the permeation of only the smaller molecules.

Multi-layer Diffusion



Figure 6: Multi-layer Diffusion (Basile, 2011)

When the molecule–surface interactions are strong multi-layer diffusion occurs. This mechanism is like to an intermediate flow regime between surface diffusion and capillary condensation.

Poiseuille (Viscous) Mechanism



Figure 7: Poiseuille (Viscous) Mechanism (Basile, 2011)

This mechanism occurs when the average pore diameter is bigger than the average free path of fluid molecules. In this case, no separation takes place.

Knudsen Mechanism



When the average pore diameter is similar to the average free path of fluid molecules, Knudsen mechanism takes place.

CHAPTER 3:

METHODOLOGY

3.1 Research Methodology and Project Activities

The methodology for conducting this research project is exploration and discovery. As this project is mainly an empirical research, the results obtained from this research can be used to compare with other literature results. The project activities in this research are mainly experimental work. After thorough literature review is done, experimental works can be conducted to investigate the effect of feed temperature and composition of propane toward the separation of CO_2 - Methane – Propane.

3.2 Experimental Procedures/Approach

The figure below shows the general experimental procedures that will be implemented in this research project.



Figure 9 : The schematic diagram depicting the general approach in this project

3.2.1 Lab scale hollow fiber membrane module

The commercial modules diameters usually vary from 1 to 12 inches. For lab-scale module, the preferred diameter is around ¹/₂ in to ³/₄ in. One of the most important parameters to measure the effects of mass and heat transfer when module sizes change is the packing density. Packing density of a hollow fiber membrane module can be defined as the fraction of the cross-section of all fibers over the cross-section area of the module, where

packing density (
$$\phi$$
)
= no. of fibers (n)
 $\times \frac{\text{cross-section area of a fiber } (A_{\text{fiber}})}{\text{cross-section area of module } (A_{\text{module}})}$

$$\phi = n \times \frac{(\text{o.d.}_{\text{fiber}})^2}{(\text{i.d.}_{\text{module}})^2}$$

Lab-scale hollow fiber membrane module with high packing density should meet these requirements:

- a) The packing density should be near to the packing density of commercial one. (Usually above 45%)
- b) The assembling and dismantling process should be as easy as possible; commercially available parts, apparatus and materials are preferred for the module construction.



Figure 10: The hollow fiber membrane module structure



Figure 11: The lab-scale hollow fiber membrane module structure

3.2.2 Potting fibers in the module

- Fiber bundle preparation
 - (a) With assumption of 20% 50% of packing density, the number of fiber and the length of fiber required are calculated based on the diameter and length of module.
 - (b) The fiber is cut into desired length. Then, placed the fibers in parallel order and placed the together as a fiber bundle.
 - (c) A piece of Parafilm M[®] is cut with a dimension of about 40 mm long and 10 mm wide. After removing the paper backing, the Parafilm is stretched to four or five times of its length. The end of the fiber bundle is wrapped with the stretched Parafilm. The ed of the fiber bundle will become denser because of the shrinkage of the film.
 - (d) The wrapped end is cut with a sharp razor to get a smooth end.
 - (e) A layer of Araldite[®] 5 Min is applied to the smooth end before the epoxy is applied.
 - (f) The smooth end is encircled with a thin string as show on Figure 6 (D).
- Module assembly
 - (a) The shell is placed vertically and leaves sufficient space under the module to accommodate the fiber bundle.
 - (b) The fiber bundle is pulled through the shell lumen and placed at designated position of the shell. The untied end of the fiber bundle is suspended freely, thus the fiber will become ordered and packed naturally when being pulled into the shell.

(c) Seal both ends of the shell with epoxy adhesive.



Figure 12: Budle preparation and module assembly



Figure 13: Epoxy adhesive

• Post treatment after casting

The solidified epoxy resin located outside the module tube sheets is removed with a hand drill and a clean cross-section is cut using a sharp operating blade (knife), clean-up residual epoxy resin on the thread area, and seal two tube sheets of the module by connectors and reducing unions.



Figure 14: Removing solidified epoxy resin using hand drill

3.2.3 Gas separation testing unit

Hollow fiber membrane module is installed in the experimental set up as shown in diagram below.



Figure 15: Flow sheet of gas separation testing unit for experimental validation.

The testing unit mainly consists of gas cylinders, mass flow controllers, compressor, and infrared analyser. Natural gas (with impurities) and pure methane can be used alternatively in the set up. In addition, nitrogen is used for purging the separation system. In the current study, pure methane, propane and CO₂ are used to evaluate the performance of separation system. Thermocouples and pressure gauges are installed before and after the permeation test cell to monitor the temperature and pressure drop across the membrane module. Furthermore, a back pressure regulator is fixed after the membrane module in order to generate trans-membrane pressure required for the separation of gases. The whole system except feed cylinders and compressor is placed in an oven to maintain the temperature of system and isolate from external effects. Coriolis flow meters are used to measure the mass flow rates of streams. Similarly, Infrared analyzer is used to measure the composition of feed, permeate and retentate streams. They are connected to data acquisition system in order to record the gas concentrations of streams at different times.

3.3 The effect of feed temperature

The first part of this project is conducted to study the effect of feed temperature to the separation of $CO_2 - CH_4 - C_3H_8$. Four sets of feed temperature values (40°C, 50 °C, 60 °C and 70 °C) were used to investigate the parameter. Each set of experiment was conducted for 3 hours. The condition of the experiments in this part is as follow:

Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO ₂ Flowrate	= 3 L/min
CH ₄ Flowrate	= 2 L/min
C ₃ H ₈ Flowrate	= 1 L/min
No of Fiber Used	= 10

3.4 The Effect of Propane composition

The second part is conducted to investigate the effect of feed temperature to the separation of $CO_2 - CH_4 - C_3H_8$. So, four sets of propane composition values (4%, 6%, 8% and 10%) are used to investigate the parameter. Time taken for each set to be conducted is 3 hours. These are the conditions of the experiment of this part:

Feed Temperature	$= 70^{\circ}C$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
No of Fiber Used	= 10

3.5 Gantt Chart

3.5.1 Final Year Project 1

No.	Detail/ Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	First meeting with coordinator and supervisors														
2	Literature review compilation and analysis														
3	Start to write the Extended proposal														
4	Submission of Extended Proposal (First Draft)														
5	Submission of Extended proposal (Final Draft)														
6	Proposal Defense														
7	Cleaning the module and potting fiber														
8	Request for CO2SMU Lab Booking for FYP 2														
9	Purchase the gas required (Propane and Pentane)														
10	Submission of Interim Draft Report														
11	Submission of Interim Report														

Completed Activities Uncompleted Activities

3.5.2 Final Year Project 2

No.	Detail/ Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	Potting the fiber into the module														
2	Experiment: Feed Temperature = 40°C														
3	Experiment: Feed Temperature = 50°C														
4	Experiment: Feed Temperature = 60°C														
5	Experiment: Feed Temperature = 70°C														
6	Experiment Propane Composition = 4 %														
7	Experiment Propane Composition = 6 %														
8	Experiment Propane Composition = 8 %														
9	Experiment Propane Composition = 10 %														
10	Start to write Progress Report														
11	Submission of Progress Report (Final Draft)														
12	Preparation for Pre-SEDEX (Poster, presentation, etc.)														
13	Pre-SEDEX														
14	Submission of Dissertation (soft bound) and Technical Paper														
15	Oral Presentation														
16	Submission of Project Dissertation (Hard Bound)														

CHAPTER 4:

RESULT AND DISCUSSION

4.1 Effect of Feed Temperature

The study of feed temperature effects on gas permeance is done by selecting feed temperature range of 40°C to 70°C and operating pressure of 9 bar.



Figure 16: Graph of Permeance vs Feed Temperature

From the result, it is observed that the permeance of CO_2 is decreasing as the feed temperature increase. The same trend occurred to methane and propane. This situation can be explained by the relationship of temperature and permeability as shown in the surface diffusion equation:

$$P_{s,i} = \frac{2\varepsilon^2 t_m (1-\varepsilon) D_{s,i} \rho_m f_i}{z\tau^2 RPT r_{p,i}}$$

Where:

 $P_{s,i}$ = is permeability of gas as a result of surface diffusion

$$\epsilon$$
 = porosity

$$\tau$$
 = tortuosity

 t_m = membrane thickness

 ρ_m = membrane density

 $f_i = loading factor$

As the temperature increase, the permeability will decrease and that is why the permeance increase when the feed temperature increases. In the graph, it is observed that at feed temperature 40°C gives the highest permeance. Therefore, it is clear that permeance will decrease as temperature increases. This statement is also supported by Boributh (2009) where it is stated that when the temperature increase will caused decreasing in flux.

Most hydrocarbon streams also contain some portion of water that will cause the CO₂ to become soluble in water as temperature increases. The water could also vaporize and block at the membrane pores, thus, inhibiting other species to pass through (Boributh, 2009).

In addition, Safari (2009) told that the relationship between the cost and the operating temperature is proportional. So, when the temperature increase, the cost will increase and this situation makes higher temperature become less attractive.

4.3 Effect of Propane Composition

Based on the result obtained from this part, the graph of permeance vs gas composition is plotted.



Figure 17: Graph of Gas Permeance vs Propane Composition

For this part, the study of different propane composition (4%, 6%, 8% and 10%) is done at operating temperature and pressure of 40°C and 9 bar respectively. The feed CO₂ concentration is kept at 50% for all sets of experiment in this part as mentioned by Cakal (2012) that CH₄ permeation detected in the gas chromatograph is very limited in CO₂ rich mixture. From the result, it is observed that as the propane composition increases the permeance of CO₂, CH₄ and C₃H₈ is decreasing. This is due to CH₄ and C₃H₈ loss to permeate side.

In higher feed propane composition, the methane and propane loss is higher due to initial abundance presence of methane and propane molecules in the treated stream as compared to CO_2 , which caused the tendency for methane and propane loss to the permeate site is higher. Therefore, the permeance of the gas is decreases.

CHAPTER 5:

CONCLUSION

This project aimed to study the effect of feed temperature and propane composition on the separation of CO_2 - CH_4 - C_3H_8 . Basically this project is divided into two parts according to the parameter of study.

The first part of the study is focusing on effect of feed temperature on the gas permeability. From the result, the gas permeability increases as the feed temperature increase.

The second part focused on the other parameter which is propane composition. Holistically, the propane composition increment will caused the gas permeabily to decrease due to CH_4 and C_3H_8 loss to permeate side.

In conclusion, the main objective of this project which is to study the effect of feed temperature and the propane composition to the separation of CO_2 - CH_4 - C_3H_8 is achieved.

REFERENCES

Abedini, R., Nezhamoghadam, A., (2010), 'Application of Membrane in Gas Separation Processes: Its Suitability and Mechanisms', Petroleum and Coal, pp 69-80

Ahmad, F., et al., (2012) Temperature and pressure dependence of membrane permeance and its effect on process economics of hollow fiber gas separation system. Journal of Membrane Science, 2013. 430(0): p. 44-55.

Basile, A., Gallucci, F., (2011), 'Membranes for Membrane Reactor: Preparation,Optimization & Selection'

Burggraaf, A.J.(1996), 'Fundamentals of Inorganic Membrane Science and Technology'.

Dutta, BK., (2007), 'Principles of Mass Transfer and Separation Process', Eastern Economy Edition, Chapter 14 Membrane Separation, pg 728.

Geankoplis, CJ., (2003), 'Transport Processes and Separation Process Principles', Prentice Hall.

Iqbal, M.,(2007) Development of Asymmetric Polycarbonate (PC) Membrane for Carbon Dioxide Removal from Methane, in Msc Thesis, University Technology of Petronas, Malaysia: Malaysia.

Jusoh, N., Lau, KK., & Shariff, AM. (2012). *Parameter Estimation in Removal of CO2* from Natural Gas Using Membrane Processes. Tronoh: UTP.

Katoh, T., et al., (2011) Dynamic simulation of multicomponent gas separation by hollow fiber membrane module:non ideal mixing flows in permeate and residue sides by using the tanks-in sereies model. Sep. Purif. Technol.

Kirk-Othmer, (2004) Membranes, Hollow-Fiber, in Kirk-Othmer Encyclopedia of Chemical Technology, John Wiley and Sons. p. 1-28.

Koros, WJ., (1996) Terminology for membranes and membrane processes. Pure and Applied Chemistry.

Mellon, N. Shariff, AM., Lau, KK., (2009), 'A Theoritical Analysis of Natural Gas Dehydration Using Membrane Processes', Department of Chemical Engineering, Universiti Teknologi PETRONAS (UTP).

Mulder, M., (1996) Basic Principles of Membrane Technology Second Edition. Kluwer Academic Publisher.

Paul, G., Lake, L., & Gould, T. (1986). A Simplified Predictive Model for CO2 Miscible Flooding.

Pontious, S. S., & Tham, M. S. (1978). North Cross (Devonian) Unit CO2 Flood -Review of Flood Performance and Numerical Simulation Model.

Prieditis, J. B. (1993). Effects of Recent Relative Permeability Data on CO2 Flood Modeling.

Rao, D. N., & Hughes, R. G. (2011). Current Research and Challenges Pertaining to CO2 Flooding and Sequestration.

S. Wo, S. a., B. Blakeney-DeJarnett, B. C., & and C.E. Mullen, E. P. (2008). Simulation Evaluation of Gravity Stable CO2 Flooding in the Muddy Reservoir at Grieve Field, Wyoming.

Saripalli, P., & McGrail, P. (2001). Semi-Analytical Approaches to Modelling Deep Well Injection of CO2 for Geological Sequestration.

Schell, W., & Houston, C. (1983). Membrane Gas Separations for Chemical Process and Energy Application, Chap 7. In *Indusrial Gas Separation* (pp. pp.125-143).

Schrier, J. (2012). *Carbon Dioxide Separation with a Two-Dimensional Polymer*. Pennsylvania, United States: ACS.

Weller, S., W.A. Steiner. (1950), 'Separation of Gases by Fractional Permeation throughMembranes', Journal of Applied Physics, Vol. 21, pp 180.

APPENDIX

Data and result obtained from the experiment:

The first part of this project is conducted to study the effect of feed temperature to the separation of $CO_2 - CH_4 - C_3H_8$. So, four sets of feed temperature values (40°C, 50 °C, 60 °C and 70 °C) are used to investigate the parameter. Each set of experiment is conducted for 3 hours. Table below show the data and composition (in vol %) of Carbon Dioxide, Methane and Propane for each sets of experiment.

Set 1:

Feed Temperature	$= 40^{\circ}$ C
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 3 L/min
CH4 Flowrate	= 2 L/min
C3H8 Flowrate	= 1 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 1 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	40	39	40	39
Flow (kg/hr)	0.32	0.63	58.80	57.00
CO2 Composition (Vol %)	58.80	30.10	39.20	39.94
CH4 Composition (Vol %)	39.20	21.09	2.00	3.06
C3H8 Composition (Vol %)	2.00	48.81	58.80	57.00

Set 2:

Feed Temperature	$= 50^{\circ}C$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 3 L/min
CH4 Flowrate	= 2 L/min
C3h8 Flowrate	= 1 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	50	39	40	39
Flow (kg/hr)	0.37	0.63	0.62	0.02
CO2 Composition (Vol %)	58.80	52.60	52.50	90.80
CH4 Composition (Vol %)	39.20	36.86	37.70	8.00
C3H8 Composition (Vol %)	2.00	10.54	9.80	1.20

Set 3:

Feed Temperature	$= 60^{\circ}\mathrm{C}$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 3 L/min
CH4 Flowrate	= 2 L/min
C3h8 Flowrate	= 1 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	60	39	40	39
Flow (kg/hr)	0.56	0.63	0.61	0.02
CO2 Composition (Vol %)	58.80	34.50	35.60	89.50
CH4 Composition (Vol %)	39.20	24.17	24.57	8.00
C3H8 Composition (Vol %)	2.00	41.33	39.83	2.50

Set 4:

$= 70^{\circ}C$
= 9 Bar
= 8 Bar
= 3 L/min
= 2 L/min
= 1 L/min
= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	70	39	40	39
Flow (kg/hr)	0.61	0.63	0.60	0.03
CO2 Composition (Vol %)	58.80	30.10	31.90	80.30
CH4 Composition (Vol %)	39.20	21.09	21.30	8.00
C3H8 Composition (Vol %)	2.00	48.81	46.80	11.70

Feed Temperature (°C)	Permeance (GPU)				
	CO_2	CH ₄	C_3H_8		
40	179.84	43.57	4.72		
50	107.9	26.14	1.51		
60	88.63	21.79	1.45		
70	53.01	14.52	0.87		

Table 1: Effect of Feed Temperature on Gas Permeance

The second part of this project is conducted to study the effect of propane composition to the separation of $CO_2 - CH_4 - C_3H_8$. So, four sets of feed temperature values (4%, 6%, 8% and 10[%]) are used to investigate the parameter. Each set of experiment is conducted for 3 hours. Table below show the data and composition (in vol %) of Carbon Dioxide, Methane and Propane for each sets of experiment. Each set of experiment is conducted for 3 hours. The condition and result obtained from this part is as follow:

Set 1:

Feed Temperature	$= 40^{\circ}\mathrm{C}$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 5 L/min
CH4 Flowrate	= 4.6 L/min
C3H8 Flowrate	= 0.4 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	40	39	40	39
Flow (kg/hr)	1.11	1.13	1.12	0.01
CO2 Composition (Vol %)	50.00	50.00	31.90	88.9
CH4 Composition (Vol %)	46.00	46.00	48.81	8.00
C3H8 Composition (Vol %)	4.00	4.00	19.29	3.10

Set 2:

Feed Temperature	$= 40^{\circ}C$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 5 L/min
CH4 Flowrate	= 4.4 L/min
C3H8 Flowrate	= 0.6 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	40	39	40	39
Flow (kg/hr)	1.11	1.13	1.12	0.01
CO2 Composition (Vol %)	50.00	50.00	35.60	86.00
CH4 Composition (Vol %)	44.00	44.00	48.13	8.00
C3H8 Composition (Vol %)	6.00	6.00	16.27	6.00

Set 3:

Feed Temperature	$= 40^{\circ}\mathrm{C}$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 5 L/min
CH4 Flowrate	= 4.2 L/min
C3H8 Flowrate	= 0.8 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	40	39	40	39
Flow (kg/hr)	1.11	1.13	1.12	0.01
CO2 Composition (Vol %)	50.00	50.00	38.6	84.1
CH4 Composition (Vol %)	42.00	42.00	45.61	8.00
C3H8 Composition (Vol %)	8.00	8.00	15.79	7.90

Set 4:

Feed Temperature	$=40^{\circ}\mathrm{C}$
Feed Pressure	= 9 Bar
Back Pressure	= 8 Bar
CO2 Flowrate	= 5 L/min
CH4 Flowrate	= 4.0 L/min
C3H8 Flowrate	= 1.0 L/min
No of Fiber	= 10

	Manifold 1 (Feed)	Manifold 2 (Feed)	Manifold 3 (Retentate)	Manifold 4 (Permeate)
Pressure (Bar)	9	9	8	0.03
Temp (°C)	40	39	40	39
Flow (kg/hr)	1.19	1.25	1.24	0.01
CO2 Composition (Vol %)	50.00	50.00	52.50	82.50
CH4 Composition (Vol %)	40.00	40.00	44.68	8.30
C3H8 Composition (Vol %)	1.00	1.00	2.82	9.20

Composition (%)			Permeance (GPU)		
CO_2	CH_4	C_3H_8	CO_2	CH_4	C_3H_8
50	46	4	179.84	43.57	4.72
50	44	6	107.9	26.14	1.51
50	42	8	88.63	21.79	1.45
50	40	10	53.01	14.52	0.87

Table 2: Effect of Propane Composition on Gas Permeance