Parametric Study of Single and Double Stage Membrane Configuration in Methane Enrichment Process

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***Abstract*— Operational study of a biogas upgrading plant with cleaning and methane (CH_4) enrichment has been presented in this study. Parametric study was conducted to investigate the effect of variation of process conditions for single stage without recycle (SSWR) and double stage with permeate recycle (DSPR) on product purity, CH_4 recovery and compression power requirement. In the study, achieving high CH_4 recovery and product purity simultaneously could not be attained in SSWR configuration. The performance of DSPR yielded a better result but with higher membrane area and compression power. DSPR configuration achieved high CH_4 recovery and purity at increasing feed pressure, selectivity and feed flow. The CH_4 losses increased in both configurations as $\%CO_2$ increased in the feed. DSPR configuration is considered the best configuration due to the end use of the product, as vehicular fuel, which requires high product purity.**

***Keywords*— Biogas, methane enrichment, parametric study, membrane configuration**

I. Introduction

TO reduce greenhouse gas emission and secure long-term energy supply globally, our exploitation of the earth's finite resources such as fossil fuel must be reduced, while renewable energy must be developed as alternatives. The proportion of energy generated from renewable resources is expected to increase to >20% by 2020 [1]. During the same period, the greenhouse gas should decrease by 14% in 2020 compared with 2005 [1]. The use of biogas is considered as one of the most efficient means of utilizing the renewable energy and reducing greenhouse gas emission. Biogas generated from the anaerobic decomposition of organic matter comprises mainly methane (CH_4), carbon dioxide (CO_2), smaller traces of acidic gases and impurities such as hydrogen sulphide (H_2S), nitrogen (N_2), water vapour (H_2O) and traces of other volatile organic gases (VOCs) [2]. The precise concentration of these gases in any particular biogas sample depends on the source of substrate and operating process conditions, however, the reported range is typically 40-70% vol. of CH_4 while CO_2 and other trace gases takes up the remaining percentage volume [2]. Upgrading biogas to fuel grade biomethane involves two major processes; cleaning and CH_4 enrichment. The cleaning of the biogas consists of removal of acidic gases and impurities, while the enrichment process is for separation of CO_2 from biogas. The removal of trace impurities and the elimination of CO_2 from the biogas, which leads to a corresponding increase in CH_4 concentration will not only increase the calorific value of the biogas, but will also reduce corrosion of internal combustion engine parts caused by acid gas components. After upgrading, the final product is referred to as biomethane, typically containing 95-99% CH_4 and 1-3% CO_2 which makes it suitable as vehicular fuel. Current technologies for cleaning of biogas and its subsequent CH_4 enrichment are physiochemical processes which can be grouped as follows [3].

- Absorption process (physical and chemical absorption)
- Hybrid solution (mixed physical and chemical solvent)
- Physical separation (adsorption on solid surface; membrane; cryogenic)

Each of the cleaning and enrichment technologies, except for cryogenic separation processes, are in operation on large scale and can deliver biomethane that meets vehicular fuel standard [4]. The use of membranes, which is the main technology of interest in this study, exhibit many advantages in comparison to other conventional method of biogas upgrading, including lower operation cost, easier maintenance and greater process flexibility, no need for chemicals, the absence of phase and temperature changes, which results in low energy requirement [2], [5], [6]. Despite these advantages, it is difficult for membrane permeation systems to compete with the more common absorption based processes such as amine scrubbing, especially when processing high volume of biogas [7]. Higher CH_4 losses generated by membrane systems increases gas processing cost [7]. However, the CH_4 lost during the upgrading process of biogas obtained from anaerobic digesters, can be used as fuel for heat generation since anaerobic digestion typically requires higher than ambient temperature for optimal operation [7].

A membrane separation process is used to split a feed mixture containing two or more species through a semi-permeable barrier where the species that moves through the barrier is called permeate and the other that does not move through is called retentate [8]. Gases can be separated on two types of membrane; dense membrane (non-porous) and porous membrane [9]. The transportation of gases through dense membrane occurs via solution diffusion while porous membrane occurs via Knudsen flow, selective adsorption/diffusion and molecular sieving [9], [10]. The

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transportation of gases through membrane takes place when a driving force is applied to the gaseous species. This driving force is mostly due to pressure difference or concentration difference across the membrane [9]. Various membrane materials are able to separate CO_2 & CH_4 and both polymeric as well as inorganic materials can be used. However, in industrial scale gas separation, only polymeric membrane materials are applied due to their low manufacturing cost when compared to inorganic materials [11]. The efficiency of a given membrane is determined by two parameters; its selectivity and the flow through the membrane [9]. The latter often denoted as the flux or permeation rate, is defined as the volume flowing through the membrane per unit area and time [9]. In other literature, the flow through the membrane has been further divided into pressure ratio, which is the ratio of feed pressure to permeate pressure across the membrane, and stage cut, which is the fraction of the feed gas that permeates the membrane [12]. The selectivity of two gases 'a' and 'b' in a membrane is defined as the permeability of gas 'a' divided by permeability of gas 'b' which coefficient, sorption or diffusion depends on the type of material that is used in the membrane [13]. In order to apply membranes on an industrial scale, large membrane area are normally required [14]. Membranes are grouped together to form a bundle, since the permeability of a single membrane is not so high [14]. The bundle of membrane is installed into a pipe called module which is the smallest practical unit of membrane area [9]. Three types of module exist; hollow fiber modules; spiral wound modules & envelope type module [11]. Hollow fiber is commonly used in industry due its high packing density, low investment and operating cost [9], [11]. However, pre-treatment process is always required when hollow fiber is used because it is very susceptible to fouling and difficult to clean [9]. Fig. 1 shows a schematic diagram of a hollow fiber membrane [14].

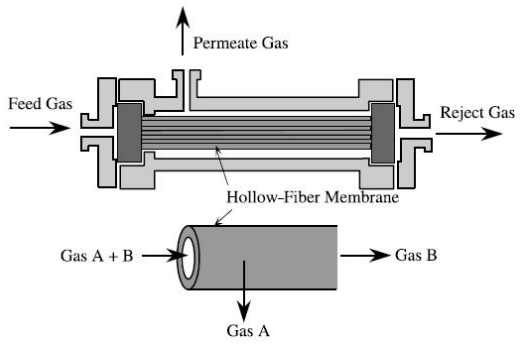

Fig. 1 Schematic diagram of a hollow fiber membrane

The arrangement of modules for separation to achieve a desired product purity and recovery of feed specie is called stage(s) [9]. This arrangement is based on economic consideration and end use of the product [9]. On economic consideration, three important elements are considered; the cost of membrane plant (membrane element and pressure housing); the capital and operating cost; and product losses [15]. The quality of the product depends on the end use. Critical operating parameters that affects the quality of upgraded biogas and CH_4 recovery in using membrane are the operating pressure, the raw biogas feed flow rate which is a function of the plant capacity, and composition of the feed [1]. Membrane configuration has been reported to be of single stage without recycle, multistep single stage and multistage configurations [3], [9], [12]. A single membrane module or a number of such modules arranged in parallel or in series without recycle with a single pass flow constitute a single stage membrane separation process [16]. The extent to which a feed mixture can be separated in the single stage is limited. Thus, in order to achieve a higher degree of separation, cascades of membrane modules with recycle are often used. Multistage configuration of two stages or more has been reported to yield higher CH_4 recovery and product purity though with higher investment cost due to additional membrane area and compression cost [15].

The usual target of an enrichment process using membrane is to produce a retentate stream essentially stripped of CO_2 and with a low flowrate but highly CO_2 rich permeate stream. These two requirements cannot be met simultaneously; a trade-off must be made between CH_4 recovery and purity. Therefore, this work presents a simulation of two possible membrane configurations of an existing operational biogas upgrading plant that uses hollow fiber membrane module for CH_4 enrichment with descriptive detail of the cleaning process. The simulation was conducted to investigate the performance of the configurations on product purity, product recovery and compressor power requirement. A parametric study investigating the effects of variations of feed composition, operating pressure and feed flow rate was conducted, with a view to investigate the performance of the configurations from an operation point of view.

II. Description of Process Plant

A. Biogas Cleaning Process

The raw biogas used in the upgrading process was generated from a closed landfill at Sebenza in the Ekurhuleni Municipality of South Africa. Pipes were bored into the landfill for extraction of the biogas generated by the natural decomposition of the organic fraction of waste. The biogas composition is as follows; CO_2 38.9%; CH_4 55%; O_2 0.4%; N_2 5%; H_2S 0.002% and H_2O 0.66%. The biogas stream was made to go through a vessel packed with high efficient iron-III-oxide (Fe_2O_3) adsorbent for preliminary desulphurization according to the following reaction:

$$Fe_2O_3.H_2O + 3H_2S = Fe_2S_3.H_2O + 3H_2O$$

The feed is at atmospheric pressure and operating temperature of 27°C. A blower was installed to increase the pressure to about 1.4bar. The temperature of the vessel was kept low to ensure the Fe_2O_3 was not dehydrated, which will reduce its reactivity. Similar recommendation for Fe_2O_3 was also given by Wang [17]. Fe_2O_3 is not regenerated in the plant, and has to be replaced once saturated with elemental sulphur. The resulting biogas stream, which was now almost completely free of H_2S, was then compressed to about 12 bar in a two stage compressor and stored temporarily in a vessel to ensure a uniform flow rate. Thereafter, the biogas was cooled to about 40°C with a heat exchange using cooling water. Possible particles and oil content in the biogas were removed by passing it through a mechanical filter and an oil separator because the compressor was oil

based for lubrication. A pressure swing adsorption (PSA) with activated alumina (Al_2O_3) was used for completed dehydration of the biogas. The N_2, siloxane, possible H_2S and other traces of volatile organic gases (VOCs) were adsorbed using an activated carbon filter. All unit operations up to this point were considered part of the cleaning process. The resulting biogas has about <5% vol. N_2, 1ppm H_2S and ppb levels for VOCs and siloxanes which is deemed not harmful to the membranes in the subsequent enrichment process.

B. Methane Enrichment

The most important task required to achieve fuel grade biomethane is the separation of CO_2 from the product gas stream. The plant studied used hollow fiber membrane for the separation of CO_2 from the biogas to achieve the desired fuel grade biomethane. The first membrane module was fed from the bore side at 10.311bar & 36°C. The permeate stream of the first membrane module which leaves at about 1.082bar was used to purge the PSA unit. The retentate stream leaves the membrane at 8.82bar and a slight temperature drop, with a high concentration of methane. This serves as the feed stream for the second stage separation. A sampling point is available for each membrane module. Gas species of the product stream can be randomly evaluated and the composition of gases through the membrane can be known. The permeate stream of the second membrane module, which contains significantly higher amounts of CH_4 as compared to permeate of the first stage, was recycled back for recompression through the suction of the compressor. Biomethane produce during the second stage separation is the desired product which is stored into a vessel after which it was odourised with mercaptan as a safety procedure and for leak detection. The last stage is compression of the biomethane to 220bar into high pressure cylinders.

III. METHODOLOGY

A. Process Simulation

The simulation of the upgrading process was done using ChemCAD, a steady state process simulator, from Chemstation. The PSA unit, a dynamic and unsteady state process, was represented with a component separator unit operation and was constrained to plant result for H_2O and H_2S removal. The cleaning process according plant data effectively eliminated vapour and H_2S content in the biogas while N_2 was reduced to acceptable limit; hence in the enrichment process simulated, CO_2 and CH_4 were only considered.

B. Data Collection

The process operating conditions were data collected from the plant. The membrane intrinsic properties (selectivity and permeability), configurations and module design data were not shared due to intellectual property infringement and confidentiality concerns. Intrinsic properties were obtained from literature while the module design data was provided by Almeesoft Engineering, USA. The module design data were verified by the plant manager and was accepted to be inconformity with industrial standard.

C. Membrane Module Design and Simulation Validation

A built-in membrane model for gas permeation available in ChemCAD 6.5.5 was used for the CH_4 enrichment. Almeesoft Engineering gas permeation software was also interface with ChemCAD during sensitivity analysis to study the effect of varying the process conditions on CH_4 recovery and product purity. The important process parameter specified for the simulation of the membrane is given in TABLE I and represents base condition for the simulation.

TABLE I
PROCESS PARAMETER FOR MEMBRANE SIMULATION

Parameter	
Feed pressure	9.9bar
Permeate pressure	1.08bar
Feed flow rate	80m^3/h
Flow mode	Counter-current
Feed side	bore
Temperature	30°C
Compressor stages	2
Cooling medium	water
CO_2 permeability[1]	440 Barrer
CH_4 permeability[1]	28.2 Barrer
N_2 permeability[1]	35.6 Barrer
O_2 permeability[1]	111 Barrer
CO_2/CH_4 selectivity	15.6
CO_2/N_2 selectivity	12.4
CH_4/N_2 selectivity	0.79
CO_2/O_2 selectivity	3.96
Account for shell pressure drop	yes

[1]Harasimowicz, et al. [18]

Two membrane configurations was considered for this study; single stage without recycle (SSWR) and double stage with permeate recycle (DSPR) as shown in Fig. 2. The configurations were simulated to investigate the performance of each configuration to varying operation parameters. The following assumptions were considered for the hollow fiber membrane module for the study. (1) Transport properties of the membrane are independent of variation in gas composition throughout the separation. (2) No flux coupling between gas components. (3) Deformation of the hollow fiber under pressure is negligible. Based on these assumptions, sensitivity analysis was carried out on both configurations and the results were discussed.

The membrane data for each module was computed based on the module specification provided by the end user into the simulator. Parameters considered in the membrane module design are as listed in the TABLE II. The process simulation for the cleaning and CH_4 enrichment process was validated by the plant upgrading process result.

TABLE II
MEMBRANE MODULE DESIGN PARAMETERS

Properties		units
Fiber internal diameter	220.98	μm
Fiber outer diameter	370.84	μm
No. of fibers	10,000	
Active length	1.1	m
Membrane area per module	11.6293	m^2
Pot length	0.0509	mm
No. of shell	1	
CO_2 viscosity	0.0153	c_p
Packing porosity	73.8%	%
Module housing	Stainless steel	

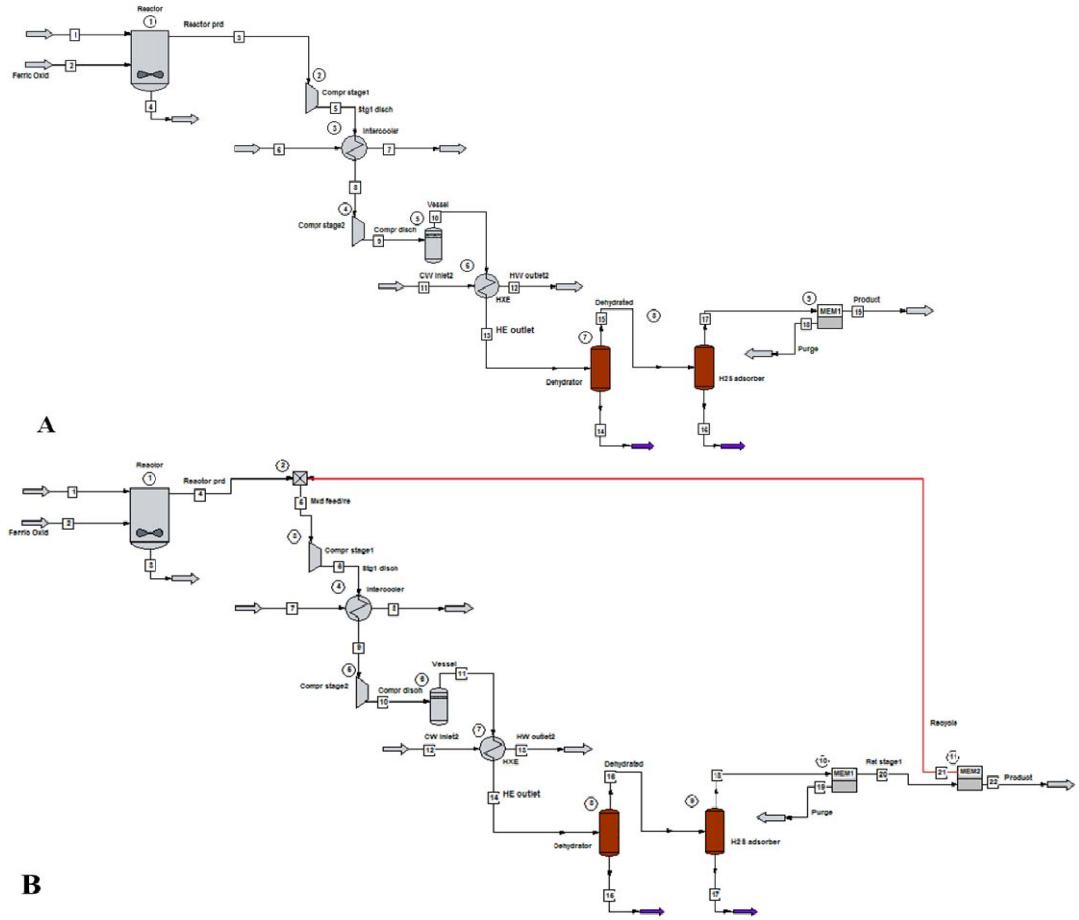

Fig. 2. Schematic diagram of the membrane configuration having (A) Single stage without recycle (SSWR), (B) Double stage with permeate recycle (DSPR).

IV. Result and Discussion

A. Process Simulation Validation

TABLE III shows the results of the simulation against those of plant upgrading. The plant had 91.16% CH_4 recovery with 87.2% CH_4 purity. The two proposed configuration for this analysis was simulated to determine their performance against plant result. SSWR yielded 87.47% CH_4 recovery with 77.89% CH_4 purity while DSPR yielded 89.01% CH_4 recovery with 84.59% CH_4 purity. The simulated DSPR process suggests a good approximation to the plant data with a percentage error of 2.64%. The error in the comparison could be attributed to higher CO_2/CH_4 selectivity of the plant membrane and feed pressure drop within the membrane module. Sensitivity study was carried out on both configurations to study the effect of varying process operating condition on CH_4 recovery, purity and compressor power requirement.

TABLE III
VALIDATION OF THE PROCESS SIMULATION WITH PLANT DATA

	Plant result			**Simulated result**						
		Final stage		SSWR		DSPR		Initial stage		Final stage
	Feed	Permeate	Retentate (Sale gas)	Permeate	Retentate	Mixed feed with recycle	Compr discharge/cleaned Feed 1	Permeate	Retentate/feed 2	Permeate to recycle	Retentate (product)
Flow (m^3/hr)	80.000	23.000	46.000	30.060	49.410	98.550	98.030	33.210	64.820	18.560	46.300
Pressure (bar)	1.000	1.082	8.280	1.080	9.400	1.000	9.900	1.500	9.230	0.800	8.769
Av. Mol. Wt.	27.606	31.847	18.384	38.501	21.079	28.890	28.949	39.594	23.496	34.426	19.111
CH_4 (% mol.)	55.000	40.120	87.200	18.351	77.892	50.660	50.934	14.657	69.518	31.947	84.590
CO_2 (% mol.)	38.934	53.997	4.900	79.201	14.854	43.626	43.862	83.265	23.677	63.843	7.563
N_2 (% mol.)	5.000	4.543	7.693	2.049	6.849	4.730	4.756	1.685	6.329	3.566	7.437
O2 (% mol.)	0.400	0.964	0.265	0.399	0.405	0.446	0.448	0.393	0.476	0.643	0.410
H_2S (% mol.)	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
H_2O (% mol.)	0.664	0.372	0.005	0.000	0.000	0.537	0.000	0.000	0.000	0.000	0.000
Heat values (BTU/SCF)											
HHV	967.4	202.9	882.0							130.4	861.2
LLV	870.9	182.7	794.0							117.3	775.3
% CH_4 Recovery			**91.16**		**87.47**						**89.01**

B. Methane Recovery and Product Purity

1) Effect of %CO_2 in Feed

Figs. 3a, 4a and 5a show the effect of increased CO_2 content in feed on CH_4 recovery, purity of product and membrane area required for separation on both configurations respectively. 89.8% CH_4 recovery was achieved with about 10% CO_2 in the feed stream in the SSWR configuration requiring 25.7m^2 of membrane area to achieve 89.85% CH_4 in the product. In DSPR configuration, CH_4 recovery was 90.48% with a product purity of 90.69% requiring 51.75m^2 membrane area for 10% CO_2 in the feed. As the CO_2 concentration increased, CH_4 recovery reduces as well as purity of product but with an increase in membrane area. With 60% CO_2 in the feed, CH_4 recovery has reduced to 84.19% with a 70.54% CH_4 in the product while for DSPR, 86.8% CH_4 recovery was achieved with 82% CH_4 in the product. Membrane area increased to 27m^2 for SSWR while DSPR required 53.58m^2. DSPR achieved higher CH_4 recovery and purity over SSWR because more membrane area was available for separation. Also the recycling of permeate of the second stage membrane separation ensures that less CH_4 is lost in DSPR configuration.

2) Effect of Feed Pressure

Figs. 3b, 4b and 5b show the effect of feed pressure on CH_4 recovery for both configurations, product purity and membrane area required respectively. The process condition and membrane shell data remains the same as earlier described except for the feed pressure been varied. It was observed as shown in Fig. 3b that increasing the feed pressure increased CH_4 recovery until a limit is reached where further increase in the pressure does not improve the CH_4 recovery rate. As the feed pressure increases from 10bar to 30bar, the CH_4 recovery increases rapidly from 93.5% to 94.96% for SSWR and thereafter the increase in pressure causes a gradual reduction in CH_4 recovery to 94.84% at 50bar. DSPR also achieved an increase in CH_4 recovery from 90.4% to 92.7% as feed pressure increased from 10-30bar. Further increase in pressure only increased the CH_4 recovery by 0.2%. CH_4 recovery was higher for SSWR when compared to DSPR because after the first stage separation, the feed pressure to the second stage which is the driving force reduces and causes a reduction in separation efficiency of the second stage as compared to the first stage. But despite a lower CH_4 recovery, the product purity achieved in the DSPR, 83.75% to 86.01%, when compared to SSWR, 77.6% to 78.8%, as shown in Fig. 3b, was much higher. At lower pressure, the high packing density advantage of hollow fiber membrane becomes increasingly important due to the high membrane area available hence higher purity can still be achieved. The increase in CH_4 recovery and purity of product can also be attributed to the fact that the increased pressure creates a greater driving force across the membrane surface area while the almost constant CH_4 recovery after its peak, in this case 30bar is due to the membrane module design.

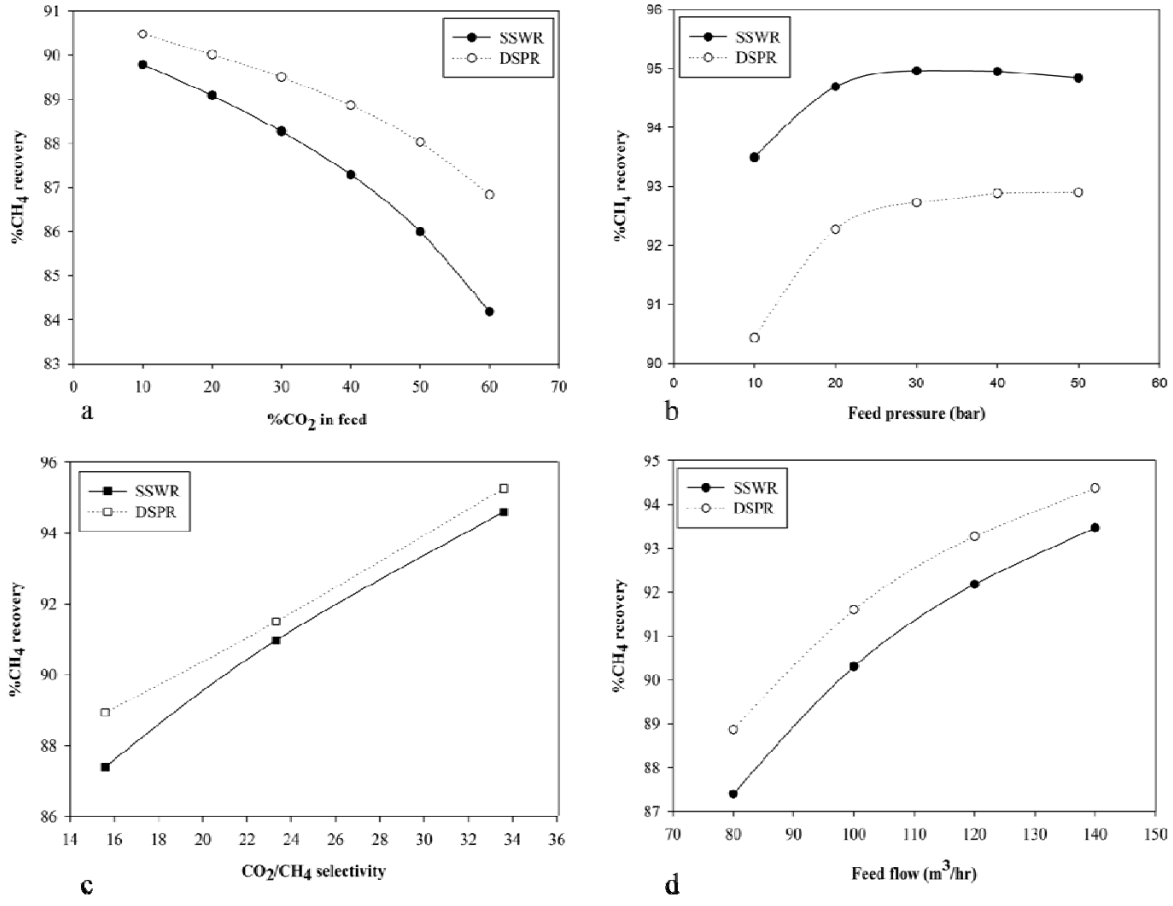

Fig. 3. Effect of variation of process parameters on CH_4 recovery

Fig. 3. (a) Effect of %CO_2 in feed, (b) Effect of feed pressure, (c) Effect of membrane selectivity, (d) Effect of feed flow rate

Improving the overall structural design will increase the membrane performance with variation in process condition [7]. This suggests that achieving high CH_4 recovery does not only depend on improving the process condition but also ensuring that an appropriate equipment design is implemented to accommodate changes in process condition. Also a higher CH_4 recovery was achieved and the required membrane area decreased as the feed pressure increases as shown in Fig. 5b. As the feed pressure increases from 10-30bar, the membrane area for SSWR decreases from 26m^2 to 9m^2 after which increasing feed pressure to 50bar only causes a reduction to 6.7m^2. DSPR uses higher membrane area for separation which ensures high product purity is achieved as feed pressure increases until it reaches its peak. The behaviour of DSPR is also similar to SSWR, as feed pressure increase to 30bar, there as a significant reduction in membrane area required from 57.6m^2 to 19.6m^2, after which an increase in pressure resulted in fluctuation in membrane area. This fluctuation suggests that the module has reached its pressure limit with respect to module design. It may be economically justified in some circumstances to increase the compression pressure of the feed stream to reduce membrane area cost and improve recovery but the structural design of the membrane casing to safely accommodate such pressure must be considered. Hence a feed pressure of 18bar will be considered as an optimal and safe operating pressure for the DSPR based on the module design data provided.

3) Effect of Membrane Selectivity

Membrane intrinsic properties have high influence on CH_4 recovery which increases with an increase in the selectivity of the membrane [3]. Figs. 3c, 4c and 5c shows the effect of selectivity on CH_4 recovery, product purity and membrane area required respectively. An increase in the membrane selectivity from 15.8 to 33.3, while other process conditions were held constant, resulted in a spontaneous increase in the CH_4 recovery from 87.4% to 94.6% for SSWR while DSPR increased from 88.9% to 95.3% as shown in Fig. 3c. The product purity for both configuration increased as selectivity increased from 15.6 to 23.3, SSWR increased from 78.3% to 79.5% while DSPR increased from 84.6% to 85.3%. A reduction was observed as the selectivity increases to 33.6 with SSWR CH_4 purity in product reducing to 76.6% and DSPR to 82.7 as shown in Fig. 4c. The reduction could be attributed to selectivity properties of the membrane towards CO_2. If less CO_2 permeate through the membrane, the CO_2 concentration in the product stream increases hence the product purity decreases. Also it has been reported in literature that highly permeable polymers have low selectivity and vice versa [19]. Hence a general trade off relationship between gas permeability and selectivity is required.

Aside increase in CH_4 recovery, increasing the selectivity reduces the required membrane area for separation as shown in Fig. 5c. With 15.6 CO_2/CH_4 selectivity, membrane area required to achieve 87.4% CH_4 recovery was 26.38m^2 for SSWR and at 33.3 selectivity, 12.63m^2 was required to achieve 94.6%CH_4 recovery. For DSPR, 52.40m^2 was required to achieve 88.9% CH_4 recovery with 84.6% product purity and at 33.6 selectivity, 27.69m^2 was required to achieve 95.3% CH_4 recovery. The decrease in membrane area is due to the intrinsic property of the membrane to permeate more CO_2 over the initial contact area of the membrane and the feed gas.

4) Feed Flow Rate

Figs. 3d and 4d shows the effect of increasing feed flow rate on CH_4 recovery and product purity. The plant been studied has the capacity to produce between 20-100m^3/hr biomethane. The base feed flow rate been used for this study was 80m^3/hr. Increasing the feed flow rate, CH_4 recovery increased from 87.4% and 88.9% at 80m^3/hr to 93.5% and 94.4% at 140m^3/hr feed flow for both SSWR and DSPR respectively as shown in Fig. 3d. The product flow rate peaked at 103m^3/hr for SSWR and 100.5m^3/hr for DSPR. This shows that if the plant is operating at full capacity, more CH_4 will be recovered in the process and CH_4 lost will be reduced. The product purity reduces as the feed flow rate increases as shown in Fig. 4d. CH_4 in product reduces from 78.27% to 69.24% for SSWR while DSPR had a reduction from 85.02% to 72.29%. It was also observed that as the feed flow rate increases from 80m^3/hr to 140m^3/hr, there was a slight increase in the membrane area. The membrane area increased from 26.38m^2 to 28.4m^2 for SSWR and from 53.2m^2 to 54.6m^2 for DSPR. The reduction in product purity as feed flow increases was due to insufficient membrane area for separation hence more CO_2 is not permeated and its presence in the retentate reduces the purity of the product.

C. Compressor Power Requirement

The effect of feed composition, feed pressure and feed flow rate on compression power required for both configurations has been investigated. Fig. 6a, 6b and 6c shows the effect of increased %CO_2 in feed, feed pressure and feed flow rate on compressor power required for separation respectively. As CO_2 increases in the feed stream from 10-60%, the compression power required to facilitate the transportation of CO_2 through the membrane surface increases from 8.89hp to 34.62hp for SSWR and 17.05hp to 53.39hp for DSPR as shown in Fig. 6a. The required power per pressure increase is the minimum driving force to ensure that CO_2 permeate through the membrane surface area available and the target CH_4 recovery was achieved. Increasing feed pressure, 10-50bar, increased the required compressor power from 22.33hp to 38.82hp for SSWR and 39.78hp to 69.68hp for DSPR as shown in Fig. 6b. As earlier discussed, increasing feed pressure reduces the required membrane area but the limit of the feed pressure depends on the membrane module design pressure capacity. Increasing feed flow rate increased the compressor power required for separation as show in Fig. 6c. As the feed flow rate increases from 80m^3/hr to 140m^3/hr, the compressor power required for SSWR increased slightly from 22.18hp to 25.22hp because there was no recycling of the permeate stream. %CO_2 in the recycled stream of DSPR increased from 60.62 to 79.18% which increased the compressor power from 37.73hp to 46.26hp. As earlier discussed, increasing CO_2 in the suction of the compressor increases the required power for membrane separation to achieve the desired CH_4 recovery.

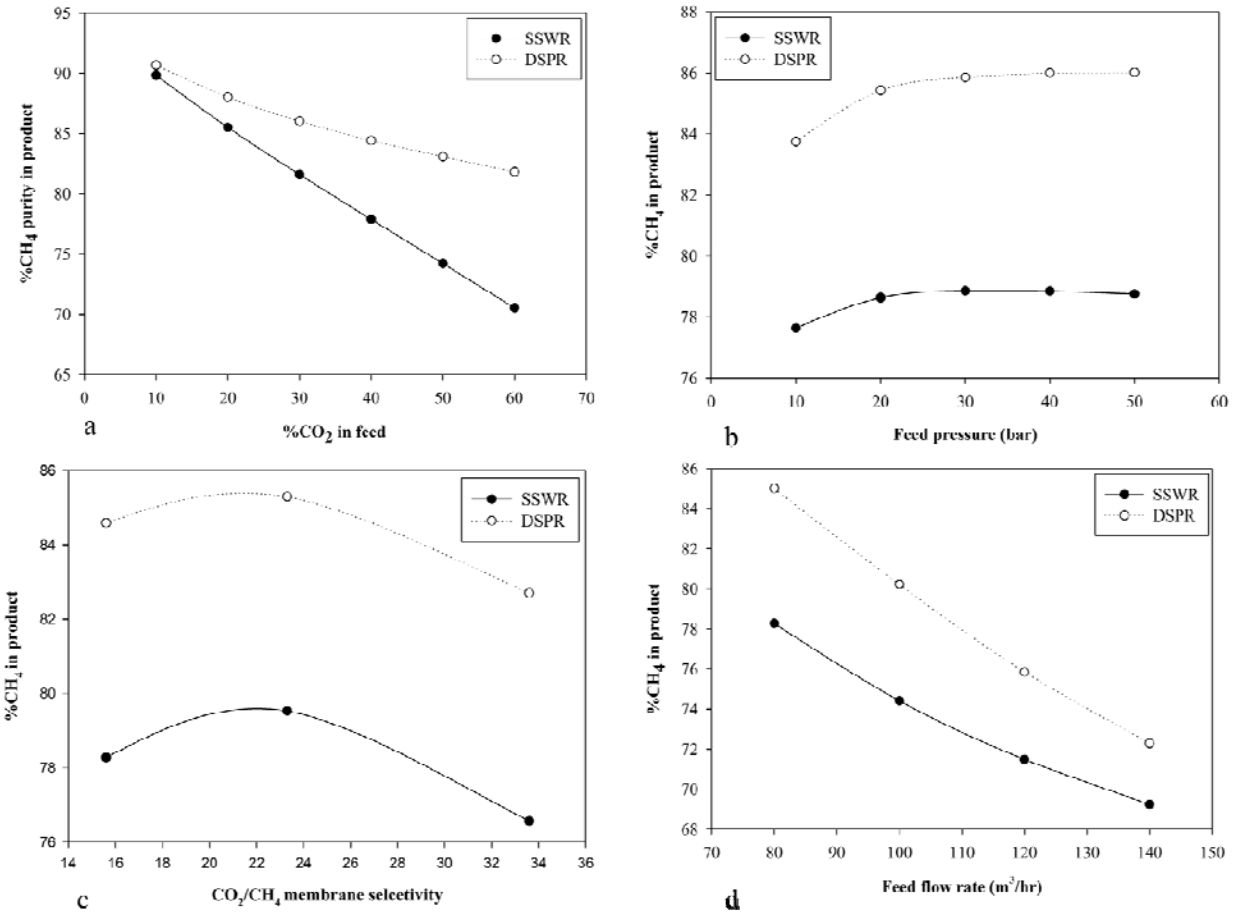

Fig. 4. Effect of varying process parameters on product purity (a) Effect of %CO_2 in feed; (b) Effect of feed pressure (c) Effect of membrane selectivity (d) Effect of feed flow

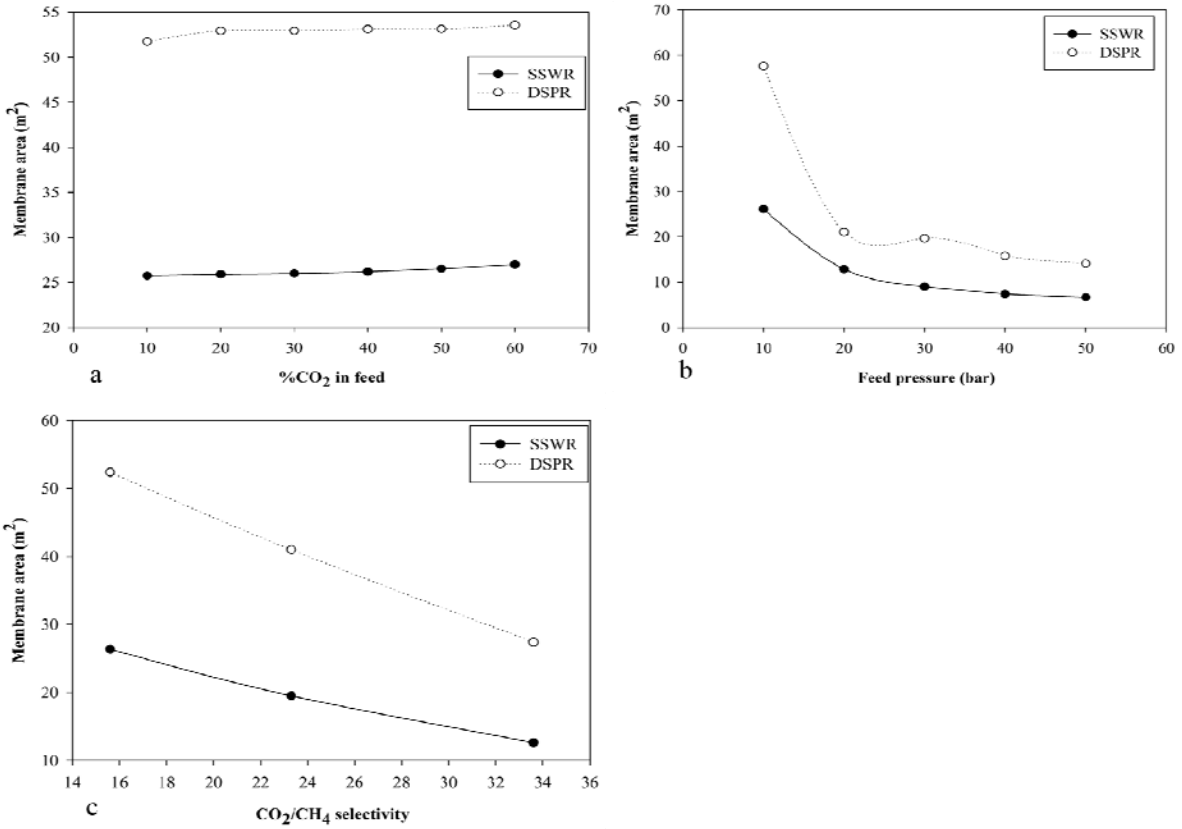

Fig. 5. Effect of variation of process variables on membrane area (a) Effect of %CO_2 in feed (b) Effect of feed pressure (c) Effect of membrane selectivity

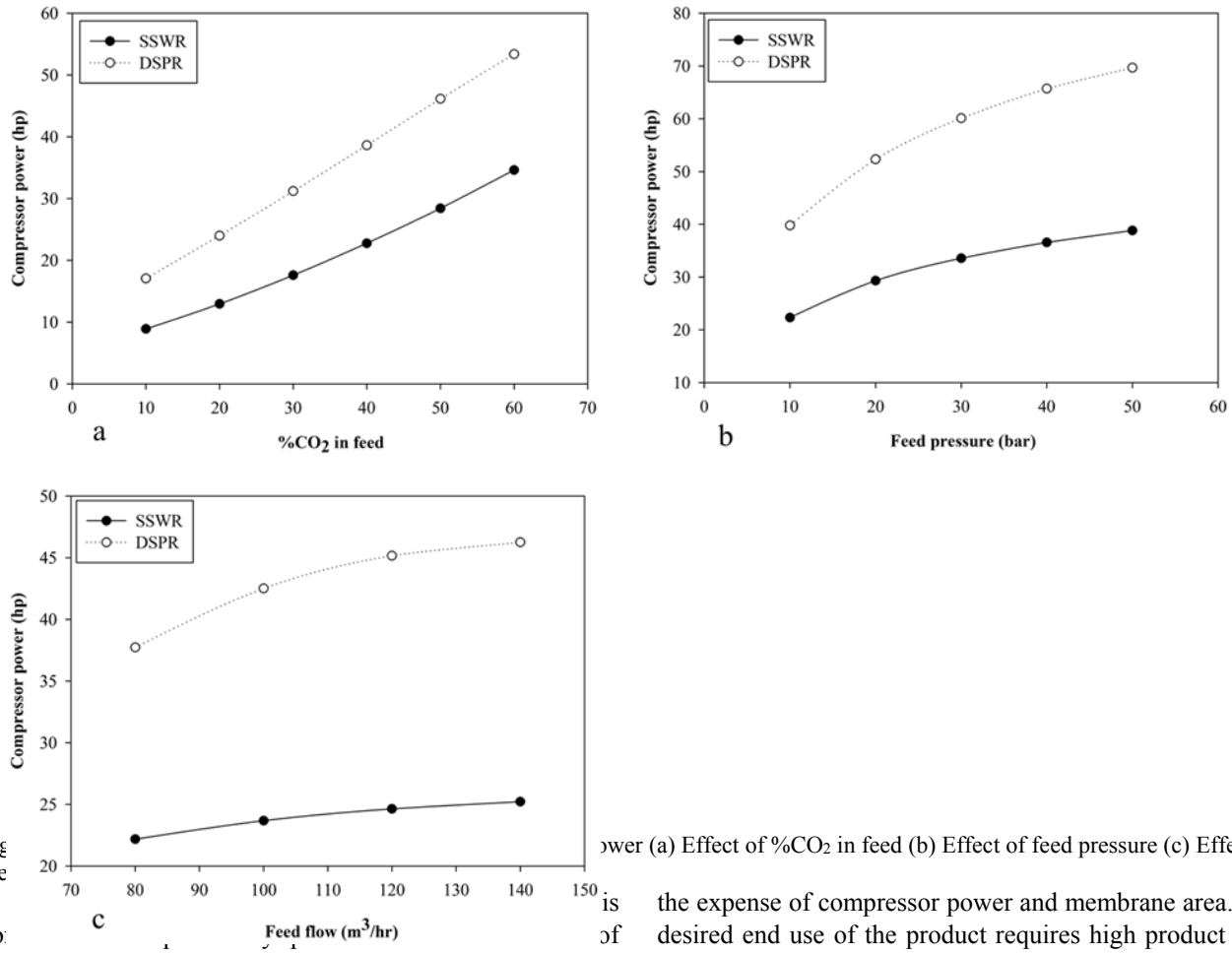

Fig ower (a) Effect of %CO_2 in feed (b) Effect of feed pressure (c) Effect of fee

is co of Deng andHagg [1], Ahmad, et al. [3] and Chmielewski, et al. [2] on membrane performance in methane enrichment process.

V. CONCLUSION

Parametric study of a biogas upgrading plant that considered cleaning of impurities in biogas and subsequent CH_4 enrichment using membrane has been presented. The simulated process was validated with an existing biogas upgrading plant result. The parametric study investigated the effect of varying feed composition, pressure and flow rate in enrichment process on CH_4 recovery on two configurations. Increasing CO_2 in feed reduces the CH_4 recovery by 6.23% for SSWR and 4% for DSPR. Product purity for SSWR reduced by 21.5% while DSPR product purity reduced by 9.7% as CO_2 increases in the feed. An increase in membrane selectivity from 5.68 to 33.33 increased CH_4 recovery by 8.2% for SSWR and a reduction in membrane area by 52.1%. Also, the increase in selectivity increased CH_4 recovery by 7.11% in DSPR and a reduction in membrane area by 47.73%. The product purity in DSPR was 8% higher than SSWR. Pressure increase also increased CH_4 recovery up to the module design configuration limit. Increasing feed flow rate from 80-140m^3/hr increased CH_4 recovery by 6.98% for SSWR. Similarly, 6.2% increase in CH_4 recovery was achieved in DSPR with a product purity that is 4.4% higher than SSWR. SSWR configuration requires less compression power and membrane area but CH_4 recovery and purity of the product stream was low. DSPR achieved higher CH_4 recovery and product purity at the expense of compressor power and membrane area. If the desired end use of the product requires high product purity and recovery like the plant under study, a DSPR is recommended though the investment cost may be high due to increased membrane area and compression cost. SSWR configuration will be suitable if product purity required is not high.

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REFERENCE

[1] L. Y. Deng and M. B. Hagg, "Techno-economic evaluation of biogas upgrading process using CO_2 facilitated transport membrane," *International Journal of Greenhouse Gas Control,* vol. 4, pp. 638-646, Jul 2010.

[2] A. G. Chmielewski, A. Urbaniak, and K. Wawryniuk, "Membrane enrichment of biogas from two-stage pilot plant using agricultural waste as a substrate," *Biomass & Bioenergy,* vol. 58, pp. 219-228, Nov 2013.

[3] F. Ahmad, K. K. Lau, A. M. Shariff, and G. Murshid, "Process simulation and optimal design of membrane separation system for CO_2 capture from natural gas," *Computers & Chemical Engineering,* vol. 36, pp. 119-128, Oct, 1 2012.

[4] E. Ryckebosch, M. Drouillon, and H. Veruaeren, "Techniques for transformation of biogas to biomethane," *Biomass & Bioenergy,* vol. 35, pp. 1633-1645, May 2011.

[5] M. H. M. Chowdhury, "Simulation, design and optimization of membrane gas separation, chemical absorption and hybrid processes for CO_2 capture," PhD, Chemical Engineering, University of Waterloo, Waterloo, Ontario, 2011.

[6] S. O. Masebinu, A. O. Aboyade, and E. Muzenda, "Enrichment of biogas for use as vehicular fuel: A review of the upgrading techniques," *International Journal of Research in Chemical, Metallurgical and Civil Engineering,* vol. 1, pp. 89-98, Jun 2013.

[7] A. Makaruk, M. Miltner, and M. Harasek, "Membrane biogas upgrading processes for the production of natural gas substitute," *Separation and Purification Technology,* vol. 74, pp. 83-92, Jul 30 2010.

[8] M. Bagajewicz, C. Ryden, and H. Baumgarner, "Use of membrane in gas conditioning," 2009.

[9] M. Mulder, *Basic principle of membrane technology*, 2 ed. Twente, Netherlands: Kluwer Academic Publisher, 1996.

[10] J. P. Williams and W. J. Koros, "Gas separation by carbon membranes," in *Advanced membrane technology & applications*, N. L. Norman, G. F. Anthony, Winston Ho, W. S., and T. Matsuura, Eds., ed Canada: Wiley, 2008, pp. 599-632.

[11] M. Scholz, T. Melin, and M. Wessling, "Transforming biogas into biomethane using membrane technology," *Renewable and Sustainable Energy Reviews,* vol. 17, pp. 199-212, Oct. 23 2012.

[12] R. W. Baker, *Membrane technology and applications*, 2 ed. Carlifornia, USA: John Wiley & Sons Ltd, 2012.

[13] F. Bauer, C. Hulteberg, T. Persson, and D. Tamm, "Biogas upgrading - Review of commercial technologies," Svenskt Gastekniskt Center (SGC) AB, Malmö, Sweden2013.

[14] Y. Kase, "Gas separation by polymide membranes," in *Advanced membrane technology & applications*, N. L. Norman, G. F. Anthony, Winston Ho, W. S., and T. Matsuura, Eds., ed Canada: Wiley, 2008, pp. 581-598.

[15] R. Spillman, "Economics of gas separation membrane processes," in *Membrane separation technology: Principles & application*, R. D. Noble and S. A. Stern, Eds., ed Beverly, USA: Elsevier, 2003, pp. 589-668.

[16] K. Scott, *Handbook of industrial membranes*. Oxford, UK: Elsevier Advanced technology, 1995.

[17] D. M. Wang, "Breakthrough behavior of H_2S removal with an iron oxide based CG-4 adsorbent in a fixed-bed reactor," Master of Science, Chemical Engineering, University of Saskatchewan, Saskatoon, 2008.

[18] M. Harasimowicz, P. Orluk, G. Zakrzewska-Trznadel, and A. G. Chmielewski, "Application of polyimide membranes for biogas purification and enrichment," *Journal of Hazardous Material,* vol. 144, pp. 698-702, Jun 18 2007.

[19] S. Kanehashi, S. Sato, and K. Nagai, "Synthesis and gas permeability of hyperbranched and cross-linked polyimide membranes," in *Membrane gas separation*, Y. Yampolskii and B. Freeman, Eds., ed United Kingdom: Wiley, 2010, pp. 3-27.