

Published in Computers and Chemical Engineering, Volume 36, 2012, Pages 119-128

<http://www.sciencedirect.com/science/article/pii/S0098135411002572>

**Process Simulation and Optimal Design of Membrane Separation System
for CO₂ Capture from Natural Gas**

F. Ahmad, *K. K. Lau, A. M. Shariff, G. Murshid

Chemical Engineering Department

Universiti Teknologi PETRONAS

Bandar Sri Iskandar, 31750, Perak, Malaysia.

***Corresponding author:** Lau Kok Keong, Chemical Engineering Department, Universiti Teknologi PETRONAS, Bandar Sri Iskandar, 31750, Perak, Malaysia.

Tel: +605-3687589 Fax: +605-3656176, E-mail: laukokkeong@petronas.com.my

Process Simulation and Optimal Design of Membrane Separation System for CO₂ Capture from Natural Gas

Abstract: In order to avoid carbon dioxide (CO₂) built up in the atmosphere, the major source of global warming, CO₂ capture must be applied to large point sources including natural gas processing. Membrane process, a relatively new technology among other available techniques, can be used for the purpose of CO₂ capture from natural gas. Over the decades, the membrane performance has been described by different mathematical models, but there is limited work done in the field of process simulation where membrane models can be incorporated with other unit operations using a commercially available simulator. In this paper, mathematical model for cross flow membrane separation has been proposed to be incorporated with ASPEN HYSYS as a user defined unit operation in order to design and optimize the membrane system for the separation of CO₂ from natural gas. The proposed simulated model is validated by published experimental and simulated data. Parameter sensitivities, along with process economics, have been studied by changing the operating conditions (feed composition and pressure) and membrane selectivity for different design configurations such as single stage (with and without recycle) and multiple stages (with permeate and retentate recycle) systems. It has been observed that double stage with permeate recycle system gives the optimum design configuration due to minimum process gas cost involved with it. The ASPEN HYSYS user defined unit operation proposed in the current paper has potential to be applied for the design, optimization and scale up of complex membrane systems.

Keywords: CO₂ capture; Membrane Process; Membrane modeling; Process simulation

Nomenclature

A_m	Total membrane area (m ²)
BPC	Base Plant cost (USD)
CC	Installed Compressor cost (USD)
CH ₄ LS	Annual cost of methane loss in permeate (USD/year)
CMC	Annual contract and material maintenance cost (USD/year)
CRC	Annual capital related cost (USD/year)
DL	Direct labor cost (USD/year)
FC	Fixed cost (USD)
GPC	Gas processing cost (USD/ MSCFD of natural gas product)
HP	Power requirement for compressors (hp)
J	Gas permeation flux through membrane (MMSCF/ft ² day)
L_f	Feed flow rate (mol/s)
L_r	Retentate flow rate (mol/s)
LOC	Annual labor overhead cost (USD/year)
LTI	Annual local tax and insurance cost (USD/year)
l	Membrane thickness (mil)
MC	Total cost of membrane modules (USD)
MMBTU	10 ⁶ BTU
MMSCFD	10 ⁶ ft ³ /day
MRC	Annual membrane replacement cost (USD/year)
MSCF	10 ³ standard cubic feet (at standard temperature and pressure)
NGLS	Annual loss of natural gas (MMSCF/year)
NHV	Heating value of natural gas (1066.8 MMBTU/MMSCF)
NWP	Wellhead price of crude natural gas (USD/MMBTU)

OSF	On stream factor
P	Project contingency (USD)
P_A	Permeability of component A (mol/MPa-m ² -s)
P_B	Permeability of component B (mol/MPa-m ² -s)
p_h	Pressure on the high pressure side (bar)
p_l	Pressure on the low pressure side (bar)
SC	Start up cost (USD)
SCF	Standard cubic feet (at standard temperature and pressure)
TFI	Total facilities investment (USD)
TPI	Total plant investment (USD)
t	Membrane life (years)
UC	Annual utility cost (USD/year)
UCP	Utility cost (USD/kwh)
VOM	Annual variable operating and maintenance cost (USD/year)
V_p	Permeate flow rate (mol/s)
x_f	Feed mole fraction
x_0	Retentate mole fraction
y_p	Permeate mole fraction

Greek Symbols:

Θ	Stage cut or fraction permeated
α	Selectivity of the membrane
Π_{cp}	Compressor efficiency (%)

1. INTRODUCTION

The increasing world-wide energy demand and pricing trends have directed the attention of oil and gas companies towards developing contaminated fields. On the other hand, global perceptions on climate change are exercising constant pressure on energy companies to adopt practices leading to the reduction of carbon emissions (Hart and Gnanendran, 2009). Therefore, CO₂ capture is attracting interest as an option for minimizing CO₂ emissions from the use of fossil fuels (Steenneveldt et al., 2006).

The amount of CO₂ contents in the natural gas can vary from 4% to 50% depending on the gas source. Before the transportation of natural gas, it must be pre-processed in order to meet the typical pipeline specification of 2%-5% CO₂ (Datta and Sen, 2006; Safari et al., 2009). Currently, many natural gas wells are undeveloped due to their low production rate and poor quality, i.e., the high CO₂ content, which has created the necessity to develop efficient processes for the separation of CO₂ from natural gas (Lee et al., 1995).

CO₂ can be removed by a number of processes considering the factors of; capital and operating costs, gas specifications and environmental concerns. The major processes can be grouped as follows:

- Absorption Processes (Chemical and Physical absorption)
- Adsorption Process (Solid Surface)
- Hybrid Solution (Mixed Physical and Chemical Solvent)
- Physical Separation (Membrane. Cryogenic Separation) (Maddox, 1982; Koros and Chern, 1987)

Membranes processes represent commercially proven technology for natural gas processing application. For a gas to permeate through a membrane surface, the gas must first

dissolve in the high-pressure side of the membrane, diffuse across the membrane wall, and evaporate from the low-pressure side. Gas separation therefore works on the principle that some gases are more soluble in, and pass more readily through polymeric membrane than other gases (Geankoplis, 2003; Baker, 2004; Ebenezer, 2005).

In the membrane process, feed gas is pretreated before entering the membrane system in order to ensure an efficient operation. It serves mainly to control the fouling, plasticization and condensation of hydrocarbons in the membranes (Baker and Lokhandwala, 2008; Xomeritakis et al., 2007). Moreover, the temperature control system is provided to maintain the gas at the desired operating temperature of the membrane fibres. Finally, the heated gas is entered into the membrane gas separators where it gets separated into two streams: the permeate, a low pressure CO₂ stream and the non-permeate or residue, a high pressure hydrocarbon rich stream (Ebenezer, 2005).

Although membrane separation technology has become a major industrial application only during the last few decades but the study of gas separation has a long history (Baker, 2004). Graham (1866) measured the permeation rates of all the gases known by that time through different diaphragms (Baker, 2004). Amerongen (1950), Barer (1951) and Stern (1966) played an important role in the development of solution diffusion model for the explanation of gas permeation (Baker, 2004). The first company to establish a Prism membrane was Monsanto that marketed for hydrogen separation, but the success of Monsanto encouraged other companies, like Cynaoi Separex and Grace Membrane Systems, to produce membrane plants for natural gas processing (Hennis et al., 1980; Baker, 2004).

Further research was conducted by Lee et al. (1994) who made field tests of membrane modules for the separation of carbon dioxide from low-quality natural gas. In their study they investigated the effects of the operating variables of pressure, feed flow rate, and the carbon

dioxide concentration in the feed. In addition to the analysis of the field data, computer models were applied for the separation of gases under perfect mixing and cross flow conditions. Lababidi et al. (1996) developed the mathematical model to optimize three configurations including single stage, two stages, and the continuous membrane column (CMC) whereas Qi and Hensen (1998) developed the optimal design strategy for spiral membrane networks for gas separations. Moreover, Qi and Hensen (2000) proposed mixed integer non linear programming for the membrane separation of multicomponent gas mixtures.

Wang (2004) enhanced the operational flexibility and adaptability of membrane process using an optimal method in which auto-controlling of the permeate gas flux was applied for the first time. Furthermore, Datta and Sen (2006) worked on the optimization of the gas processing cost for a membrane unit demonstrating that the optimum configuration might be unique within the certain ranges of CO₂ concentration and the minimum gas processing cost could only be achieved by adjusting the number of modules in each stage and the compressor power. The permeability and selectivity variations of the CO₂/CH₄ system, which included both temperature and pressure effects simultaneously, have been thoroughly studied by Safari et al. (2008). In addition, Hau et al. (2008) have investigated process design, economics, and sensitivity of the membrane stage with recycle streams.

The current paper provides analysis of different design parameters for membrane gas separation under different configurations using ASPEN HYSYS software. As membrane unit is not a pre-defined unit operation in ASPEN HYSYS, a cross flow model is proposed to predict the membrane performance in the CO₂ separation from natural gas. Finally, the proposed model is included in the process simulation as user defined unit operation along with other available unit operations. The main purpose is to optimize the membrane system configuration for

different feed compositions (from low CO₂ to high CO₂ contents), feed pressures (from low to very high pressure) and membrane selectivity.

2. METHODOLOGY

2.1. Transport Mechanism

The solution diffusion model is the most widely accepted transport mechanism for gas separation through polymer membranes. According to this model, permeants dissolve in the membrane material and then diffuse through the membrane with a concentration gradient. A separation is achieved as a result of difference in the amounts of material that dissolves in the membrane and the rate of material diffusion through the membrane. In a gas separation, a mixture of gases at a pressure p_0 is applied to the feed side of the membrane, while the permeate gas at is removed a lower pressure p_1 from the downstream side of the membrane. (Wijmans and Baker, 1995). The governing flux equation (Eq. (i)) is given by Fick's law of diffusion where driving force is partial pressure difference over the membrane.

$$\frac{q_{p,i}}{A_m} = \frac{q_p y_{p,i}}{A_m} = J_i = \frac{P_i}{l} (p_h x_i - p_l y_i) \quad (i)$$

Where J ($m^3(\text{STP})/m^2h$) is the flux of gas component i , q_p is the volume of the permeating gas (i) ($m^3(\text{STP})/h$), P_i is the permeability of gas component i ($(m^3(\text{STP})/m^2h.bar)$), P_h and P_l are feed and permeate side pressures (bar), x_i and y_i are the fractions of component i on the feed and permeate sides and A_m (m^2) is the membrane area required for the permeation.(Hussain and Hagg, 2010). The permeability (P) can be expressed as

$$P = D_{AB} \cdot S \quad (ii)$$

Where D_{AB} (m^2/s) is the diffusivity and S ($m^3(STP)/m^3.bar$) is the solubility coefficient for the gas in the membrane. The ratio of pure gas permeabilities (P_A, P_B) gives the separation factor or membrane selectivity, α (Geankoplis, 2003).

$$\alpha = P_A / P_B \quad (iii)$$

It is important to mention here that Eq. (i) can be used to accurately and predictably rationalize the properties of gas permeation membranes (Wijmans and Baker, 1995).

2.2. Process Simulation Method

In this work, a cross flow isothermal model (Figure 1) has been implemented to describe the performance of a membrane module. The model is then interfaced with Process simulation programme, ASPEN HYSYS in order to calculate permeate and retentate of the system with any number of modules, allowing complex process simulations. The programme has the possibility to use ASPEN HYSYS capabilities to calculate mass and energy balances and combine in the process model. The important process parameters are flow rates, temperatures, compositions, pressure ratio (between the upstream pressure p_h and downstream pressure p_l over the membrane) and stage cut (ratio of permeate V_p to feed flow rate L_f). The recovery of desired component (methane in our work) is calculated as

$$R = \theta \cdot y_i / x_i \quad (iv)$$

Where θ is the stage cut and x_i and y_i are feed and permeate fractions of the desired component respectively.

The model assumes no mixing in the permeate side as well as on the high pressure side. Thus, the composition of permeate can be determined at any point along the membrane by the relative

permeation rates of feed component at that point. In addition, the assumptions that follow the suggested model are:

1. It holds only for the binary gas mixture.
2. The feed side pressure of each stage is equal to the pressure of the feed stream.
3. Membrane permeability is independent of the pressure and temperature.
4. The feed side pressure drop is negligible for each stage.
5. The system operates at isothermal conditions.

For a binary gas mixture, the local permeation rate at any point in the stage over a differential membrane area dA_m , as shown in figure 1, is as follows

$$y dV = \frac{P_A}{t} [p_h x - p_l y] \quad (v)$$

$$(1 - y) dV = \frac{P_B}{t} [p_h (1 - x) - p_l (1 - y)] \quad (vi)$$

Dividing eq (i) by eq (ii), we get

$$\frac{y}{1-y} = \frac{\alpha [x - (p_l/p_h) y]}{(1-x) - (p_l/p_h) (1-y)}$$

(vii)

Where P_A and P_B are the permeabilities of pure gas components (CO_2 and CH_4 in this work), x and y are the feed and permeate composition at any point along the membrane, t is the membrane thickness and α is the membrane selectivity (Geankoplis, 2003). The above set of differential equations were solved using VB code which is used as a subroutine in ASPEN HYSYS in order to calculate the purity (permeate mole fraction y_p), methane (CH_4) recovery, and membrane area A_m required for the separation. These parameters, along with compressor

power, define the gas processing cost (GPC) for the membrane system. In order to get the optimal design, it must be minimized keeping operating conditions under consideration.

2.3. Design Configurations

The design of a membrane separation process depends on the operating parameters of the individual permeators as well as the configuration of permeators (Qi and Hensen, 1998a).. For moderate purity and recovery requirement, a single stage system, with and without recycle is appropriate (Schell and Houston, 1982). For more demanding separations, a multiple stage system is required that can further be modified by recycling permeate or retentate stream (Spillman et al., 1988; Coady and Davis, 1982).

The design of multiple stage system is very complex as it is not feasible to consider all possible configurations (Koros and Chern, 1987; Spillman, 1989; Qi and Hensen, 1998a). The conventional approach is to select a small number of design configurations and optimize the operating conditions of each configuration. The final optimum design is chosen to be the system with most favourable economics (Spillman et al, 1988; Babcock et al., 1988; Bhide and Stern, 1993; Qi and Hensen, 1998a).

The proposed design configurations include single stage (SS), single stage with permeate recycle (SSPR), double stage with permeate recycle (DDPR), double stage with retentate recycle (DDRR), triple stage with retentate recycle (TTRR) and triple stage with permeate and retentate recycle (TTPRR) as shown in Fig. 2a-2f. A compressor is needed at the start of each configuration in order to achieve the desired pressure for the parametric investigation. In addition, a smaller compressor is required to increase the pressure of permeate stream before recycling. The cooler is needed after each compression stage in order to decrease the temperature of feed stream to avoid membrane damage.

2.4. Process Conditions and Economic Parameters

The composition, flow rates, pressures and temperature of crude natural gas depend mainly on the source therefore feed conditions that are typical for medium sized natural gas treatment plant are selected. As a result, the feed flow rate of crude natural gas is maintained at 35 MMSCF (Hao et al, 2002). On the other hand, a wide range of feed pressures (10-100 bar) and membrane selectivity (5-80) has been investigated. The outlet residue CO₂ concentration is set to 2 % while outlet permeate pressure for each stage is not greater than 4 bar (Qi and Hensen, 1998a). The thickness of membrane is considered to be 1000 Å (3.937 * 10⁻⁶ in) (Hao et al, 2002). In addition, it is assumed that maximum outlet temperatures in the compressors is limited to 150⁰C giving the compression ratio of 3.5 over each compressor stage (Hussain and Hagg, 2010).

The processing cost for natural gas is often expressed as the cost per MSCF of feed but it is more reasonable to define the processing cost per MSCF of product due to presence of substantial amount of CO₂ in the feed. In addition, upgraded natural gas is sold on the basis of product volume rather than of feed volume (Hao et al, 2002). Therefore, processing cost per MSCF of product is used in the present study.

The procedure to calculate the gas processing cost (GPC) is given in Table 1. It includes the capital related cost (CRC), the variable operating and maintenance cost (VOM) and the cost of CH₄ lost in the permeate stream (CH₄LS) (Hao et al, 2008; Hussain and Hagg, 2010). The cost of cooling system is included in the compressor cost (CC) as it usually comes along with compressors. A payout time is considered to be 5 years in order to calculate the capital cost whereas project contingency, that covers the unpredictable elements of the project, is assumed to be 20% of the base plant cost (Hao et al, 2008).

3. RESULTS AND DISCUSSIONS

3.1. Model Validation

The suggested simulated mathematical model is validated by published experimental and simulated data for CO₂ removal from natural gas using membrane separation processes. The data by Pan (1986) is based on the experiments performed on sour natural gas. The feed gas used in the experiments contains 48.5 % CO₂ that is removed in the permeate stream in order to increase the recovery of methane in the retentate stream. The temperature and pressure values of the gas are 10°C and 35.28 bars, respectively, while the permeate pressure is 9.28 bar. The selectivity is assumed to be 25. The same process conditions are maintained for the proposed simulated model and compared with experimental data by Pan (1986). Table 2 shows that the suggested model gives good approximation to the experimental data with maximum percentage error < 7 %. The small error in the comparison could be attributed to the sensitivity of membrane permeability towards high pressure, which is assumed negligible in the suggested mathematical model.

The proposed simulated model is further validated by the data from Qi and Hensen (1998a) based on the study conducted on CO₂ separations from natural gas using approximate algebraic permeator model. The operating conditions and requirements are maintained in such a way to match that of simulated data by Qi and Hensen (1998a). The CO₂ concentration in the feed gas is set to 20% while permeate pressure is not allowed to exceed than 10.5 bar in order to avoid negative pressure operation. The temperature of feed gas is 40⁰C and the selectivity of membrane is considered to be 20. Furthermore, natural gas processing capacity is maintained at 19353 m³/day (6.8*10⁵ ft³/day) for the simplest design configuration of single stage membrane system without any recycle stream. Table 3 shows that the simulated model

proposed in our work is in close agreement with the simulated data by Qi and Hensen with maximum percentage error < 5 %.

3.2. Parametric and Economic Analysis

3.2.1. Methane Recovery:

Methane (CH₄) recovery decreases with the increase in CO₂ contents of the feed (Qi and Hensen, 1998). At the same time, CH₄ recovery can be improved by recycling the permeate stream as well as by using multiple stage configuration (Schell and Houston, 1982; Babcock et al., 1988; Spillman et al., 1988).

Fig. 3 shows the effect of feed composition on CH₄ recovery for all proposed configurations, for the stage cut of 0.5 and selectivity of 25. The permeability of CH₄ is considered as 1.4×10^{-3} mol/MPa-m²-s while the feed pressure and permeate pressure are maintained at 100 and 4 bar, respectively. It can be observed that the CH₄ recovery is reducing with the increase of CO₂ in the feed gas. The system without recycle, as expected, provides the lowest methane recovery. It can also be observed that methane recovery for single stage system with recycle is almost same as that of single stage system without recycle so it is not favourable to recycle the permeate stream in single stage system. Besides, the simulated results also show that the usage of a multiple stage systems leads to high methane recovery. In addition, it can be deduced that methane recovery can be improved by recycling permeate and retentate stream in the case of multiple stage systems.

Fig. 4 shows the effect of feed pressure on CH₄ recovery for different configurations. The stage cut and selectivity is same as in the previous case, whereas the feed gas contains 20% CO₂ and 80% CH₄. It can be observed that the increase in feed pressure improves CH₄ recovery. It is due to the fact that the increased pressure creates a greater driving force across the

membrane. As a result, a net increase in permeation through the membrane increases methane recovery.

Membrane properties have a high influence on CH₄ recovery, which increases with the increase in selectivity of the membrane. The reason is that increased selectivity leads to higher permeation and thus to an improved methane recovery. Fig. 5 shows the effect of membrane selectivity on the three proposed configurations. As expected, the increase in selectivity increases CH₄ recovery, especially for the multiple stage configurations. On the other hand, the increment in selectivity for the single stage configurations (with and without recycle) is less significant on the methane recovery, especially for higher selectivity membranes.

3.2.2. *Total membrane Area*

The effect of feed composition on the total membrane area required for the effective separation is studied for proposed design configurations as shown in Fig 6. It is observed that the total membrane area increases with the increase in CO₂ composition of the feed until it reaches its maximum point. After that, a further increase can lead to the decrease in the membrane area requirement. It is due to the characteristics of chosen selectivity of the membrane. It can also be observed that recycling the retentate stream in the multiple stage configurations can lead to large requirements of area, while in the single stage system, recycling has minimal effect.

On the other hand, an increase in feed pressure would decrease the total membrane area required for the effective separation as shown in the Fig. 7. It is obvious that high pressure leads to a high rate of permeation, due to which less membrane area is required for the separation.

Fig 8. shows the effect of membrane selectivity on the total membrane area for different design configurations. Increasing selectivity decreases the membrane area requirements, which is more pronounced in the multiple stage configurations, followed by single stage configuration with recycle and single stage configuration without recycle.

3.2.3. Gas Processing Cost:

In order to find the optimal design, gas processing cost (GPC) must be minimum subject to operating conditions, material and energy balances, and individual permeator mathematical model (Qi and Hensen, 1998a). The effect of feed composition on the GPC for the proposed design configurations is shown in Fig. 9. It can be noted that single stage systems without recycle (SS) yield a moderate GPC because the membrane area required is small and no compressors are needed. The double stage with retentate recycle (DSRR) and triple stage with permeate and retentate recycle (TSPRR) has the maximum GPC even in the presence of high methane recovery due to the high compressor power and very large membrane area required. Triple stage with retentate recycle (TSRR) yields relatively less GPC due to improved methane recovery. The minimum GPC is achieved by the double stage system with permeate recycle because of the high methane recovery and moderate power requirement for the configuration.

Moreover, it can also be observed that the increase of CO₂ contents in the feed gas leads to high GPC until a certain optimum point, depending upon the operating conditions and the membrane properties. The further increase of CO₂ contents can lead to the decrease in the GPC. It is due to the similar behaviour of total membrane area requirement with the change in CO₂ contents of the feed (discussed in section 3.2.2).

The effect of feed pressure on the GPC is shown in the Fig. 10. The increase in feed pressure improves the GPC, especially at relatively lower pressure ranges, due to the

improvement in methane recovery and less membrane area required for the separation at increased feed pressure.

The effect of selectivity on GPC is shown in Fig. 11. It can be deduced that GPC decreases significantly with the increase in selectivity of the membrane, which can be explained by the fact that membranes with high selectivity yield high methane recovery with less requirements of membrane area. These results are consistent with those obtained by Spillman et al. (1988), Babcock et al. (1988) and Qi and Hensen (1998a).

It can be observed in GPC comparison for different design configurations that the optimal design for CO₂ capture from natural gas using membrane is double stage with permeate recycle as it gives minimum GPC due to high methane recovery and less requirement of membrane area (even for feed with high contents of CO₂).

4. CONCLUSIONS

A systematic design strategy has been presented for the CO₂ capture from natural gas using a membrane process. The proposed cross flow model is included in the process simulation (Aspen HYSYS) as a user defined unit operation along with other available unit operations in order to design the membrane system configuration. The simulated model is validated with experimental data, where the simulated data exhibit good agreement with the published results. The design sensitivity has been investigated by changing the operating conditions and the membrane properties. It is observed that gas processing cost (GPC) increases with the increase in composition of CO₂ in natural gas to a certain point. After that, further increase in CO₂ contents can lead to the minimization of GPC. Moreover, it can be minimized by the increase in feed pressure or selectivity of the membrane. Different configurations including single stage (with and without recycle) and multiple stage membrane

systems (with permeate and retentate recycle) have been investigated for optimal design under the present study. The findings indicate that the GPC is minimum for double stage membrane system with permeate recycle making it the optimal design for the membrane separation system. Furthermore, the Aspen HYSYS user defined unit operation has potential to be applied for complex membrane system design and optimization study.

Acknowledgements

This work was done with the financial support from Universiti Teknologi PETRONOS.

REFERENCES

Amerongen GJ. Influence of structure of Elastomers on their permeability to gases. *J. Appl. Poly. Sci.* 1950; 5: 307.

- Babcock RE, Spillman, RW, Goddin, CS, Cooley TE. Natural gas cleanup: a comparison of membrane and amine treatment processes. *Energy Prog.* 1988; 8: 135–42.
- Baker RW. *Membrane Technology and Application*, 2nd ed., John Wiley & Sons, Chichester, UK; 2004.
- Baker RW, Lokhandwala K. Natural gas processing with membranes: An overview. *Ind. Eng. Chem. Res.* 2008;4: 2109-202.
- Barrer RM. *Diffusion in and through solids*. Cambridge University Press, London; 1951.
- Bhide BD, Stern SA. Membrane processes for the removal of acid gases from natural gas. I. Process configuration and optimization of operating conditions. *J. Membrane Sci.* 1993; 81: 209–237.
- Coady AB, Davis JA. CO₂ recovery by gas permeation. *Chem. Eng. Prog.* 1982; 47: 44-9.
- Datta AK, Sen PK. Optimization of membrane unit for removing carbon dioxide from natural gas. *Journal of Membrane Science* 2006; 283(1-2): 291-300.
- Ebenezer SA. Removal of Carbon dioxide from natural gas for LPG production. Semester project work. Institute of Petroleum Technology, Norwegian University of Science & Technology, Trondheim, Norway. 2006.
- Hart A and Gnanendran N. Cryogenic CO₂ capture in natural gas. *Energy Procedia* 2009;1(1): 697-706.
- Hao J, Rice PA, Stern SA. Upgrading low quality natural gas with H₂S and CO₂ selective polymer membranes Part I. Process design and economics of membrane stages without recycle streams. *Journal of Membrane Science* 2002; 209: 177-206.
- Hao J, Rice PA, Stern SA. Upgrading low quality natural gas with H₂S and CO₂ selective polymer membranes Part II. Process design, economics, and sensitive study of membrane stages with recycle streams. *Journal of Membrane Science* 2008; 320: 108-22.
- Hennis JMS, Tripodi MK. A novel approach to gas separations using composite hollow fibre membranes. *Sep. Sci. and Tech.* 1980; 15: 1059.
- Hussain A, Hagg M-B. A feasibility study of CO₂ capture from flue gas by a facilitated transport membrane. *Journal of Membrane Science* 2010;359: 140-8.
- Geankopolis CJ. *Transport processes and separation process principles*. fourth edition, Prentice Hall, New Jersey; 2003.
- Graham T. On the absorption and dialytic separation of gases by colloid septa, *Philos* 1866: 401.
- Koros WJ, Chern RT. Separation of gaseous mixtures with polymer membranes. In: Rousseau RW (Ed.), *Handbook of separation process technology*, chap 20. Wiley, New York, 1987.
- Lababidi H, Ghazi A., Al-Enezi, Ettoney HM. Optimization of module configuration in membrane gas separation. *Journal of membrane Science* 1996;112: 185-97.
- Lee AL, Feldkirchner HL, Stern SA, Houde AY, Gamez JP, Meyer HS. Field tests of membrane modules for the separation of carbon dioxide from low-quality natural gas. *Gas Separation & Purification* 1995; 9(1): 35-43.
- Liu L, Chakma A, Feng X. Propylene separation from nitrogen by poly (ether block amide) composite membranes. *Journal of membrane science* 2006; 279: 645-54.
- Maddox RN. *Gas conditioning and processing – Advanced techniques and applications*, Ed.: Campbell JM. Campbell Petroleum Series, Norman, Okla., 1982.
- Mulder M. *Basic principles of membrane technology*. 2nd edition, John Wiley, New York; 1996.

- Pan CY. Gas Separation by high flux, asymmetric hollow fiber membrane. *AIChE Journal* 1986;32: 2020-27.
- Peters MS, Timmerhaus KD. *Plant design and economics for chemical engineers*. McGraw-Hill, New York; 1980.
- Qi R, Hensen MA. Optimal design of spiral wound membrane networks for gas separations. *Journal of Membrane Science* 1998;148: 71-89.
- Qi R, Hensen MA. Optimization –based design of spiral wound membrane systems for CO₂/CH₄ separations. *Separation and Purification Technology* 1998(a);13: 209-25
- Qi R, Hensen MA. Membrane system design for multicomponent gas mixtures via mixed-integer nonlinear programming. *Computers and Chemical Engineering* 2000; 24: 2719–37.
- Safari MH, Ghanizadeh A, Montazer-Rahamti MM. Optimization of membrane based CO₂- removal from natural gas using simple models considering both pressure and temperature effects. *International Journal of Green House Control* 2008;105: 3-10.
- Shoemaker DP, Garland CW, Nibler JW. *Experiments in physical chemistry*. 6th edition, McGraw Hill, New York; 1996.
- Schell WJ, Houston CD. Spiral-wound permeators for purification and recovery. *Chem. Eng. Prog.* 1982;13: 33-7.
- Spillman RW, Barrett MG, Cooley TE. Gas membrane process optimization. In *AIChE National Meeting*, New Orleans, LA; 1988.
- Spillman RW. Economics of gas separation membranes, *Chem. Eng. Prog.* 1989; 85: 41-62.
- Steeneveldt R, Berger B, Torp TA. CO₂ Capture and Storage: Closing the Knowing-Doing Gap. *Chemical Engineering Research and Design* 2006; 84(9): 739-63.
- Wang L, Shao C, Wang H. Operation optimization of a membrane separation process through auto-controlling the permeate gas flux. *Sep. Purif. Technol.* 2007;55: 30.
- Wijmans JG, Baker RW. The solution diffusion mode: a review. *Journal of Membrane Science* 1995;107: 1-21.
- Xomeritakis G, Liu NG, Chen Z, Jiang YB, Köhn R, Johnson PE et al. Anodic alumina supported dual-layer microporous silica membranes. *Journal of Membrane Science* 2007;287(2): 157-61.

LISTS OF TABLES

Table 1: Economics parameters for gas processing cost (Hao et al., 2008)

Total Plant Investment (TPI):	$TPI = TFI + SC$
Membrane module cost (MC)	\$ 5/ft ³
Installed compressor cost (CC)	\$ 8650 * (HP/η _{cp}) ^{0.82}
Fixed cost (FC)	MC + CC
Base Plant Cost (BPC)	1.12 * FC
Project Contingency (PC)	0.20 * BPC
Total facilities investment (TFI)	BPC + PC
Start up cost (SC)	0.10 * VOM
Annual Variable Operating and Maintenance Cost (VOM):	$VOM = CMC + LTI + DL + LOC + MRC + UC$
Contract and Material Maintenance Cost (CMC)	0.05 * TFI
Local Taxes and insurance (LTI)	0.015 * TFI
Direct Labor cost (DL)	\$ 15/h
Labor Overhead Cost (LOC)	1.15 * DL
Membrane Replacement Costs (MRC)	\$ 3/ft ² of membrane
Utility Cost (UC)	\$ 0.07/kwh
Annual Cost of CH₄ Lost in Permeate (CH₄LS):	$CH_4LS = NGLS * NHV * NWP$
Annual Natural Gas Lost (NGLS)	$NGLS = 365 * OSF * L_f * y_{P(CH_4)} * x_{f(CH_4)}$
Gas Processing Cost (GPC)	$GPC = (CRC + CH_4LS + VOM) / [365 * OSF * L_f * (1 - SCE) * 1000$
Annual Capital Related Cost (CRC)	0.2 * TPI
Membrane Life (t)	4 years
Wellhead Price of Crude Natural Gas	\$ 2/MMBTU
Heating Value Of Natural Gas	1066.8 MMBTU/ MMSCF
On stream factor (OSF)	96 %
Compressor Efficiency (η _{cp})	0.8

Table 2: Validation of mathematical model with published experimental data by Pan (2008)

Stage Cut (θ)	Permeate mole fraction, CO ₂
---------------	-----------------------------------------

	Proposed simulated model	Experimental data by Pan (2008)	% Error
0.40	0.93	0.96	3.22
0.42	0.91	0.95	4.39
0.45	0.90	0.94	4.44
0.47	0.89	0.93	4.49
0.50	0.87	0.93	6.89

Table 3: Validation of mathematical model with simulated data by Qi and Hensen (1998a)

Feed Pressure (bar)	Methane Recovery (%)		
	Proposed Simulated Model	Simulated data by Qi and Hensen (1998a)	% Error
20	81	77	4.93
40	83	80	3.61
60	85	81	4.70
80	86	82	4.65
90	86	82	4.65

LISTS OF FIGURES

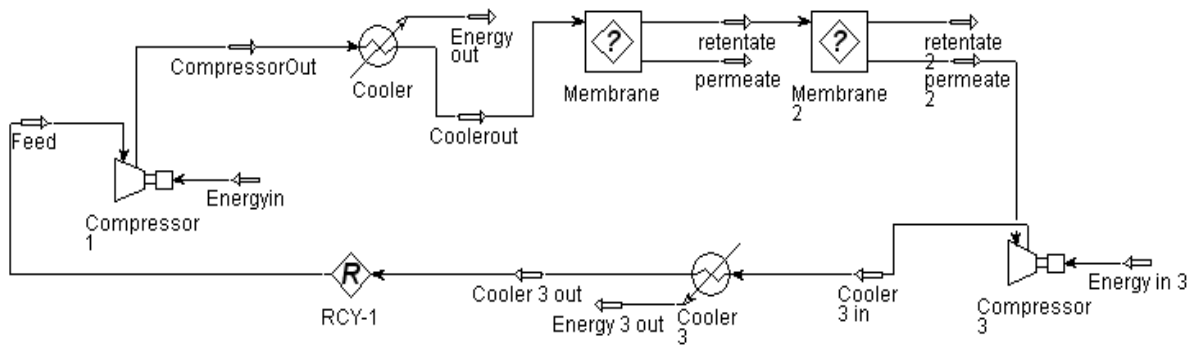


Fig. 2c. Process flow diagram (PFD) in ASPEN HYSYS for design configuration having double stage with permeate recycle (DSPR)

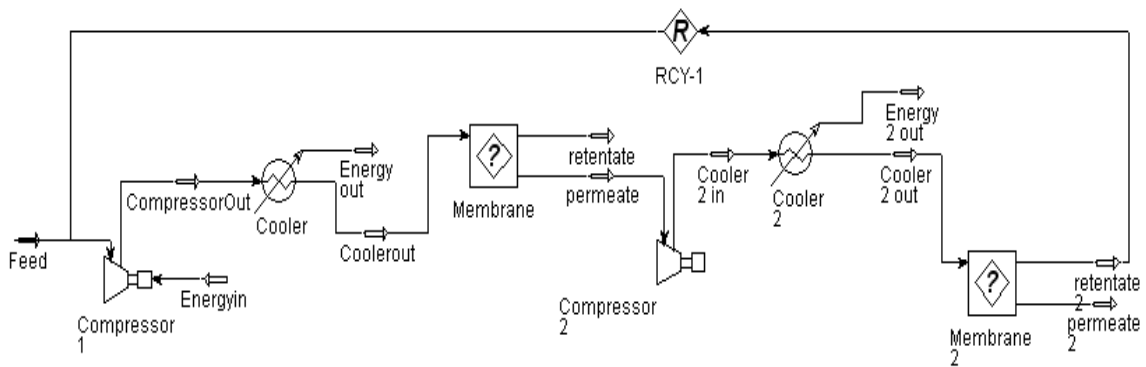


Fig. 2d. Process flow diagram (PFD) in ASPEN HYSYS for design configuration having double stage with retentate recycle (DSRR)

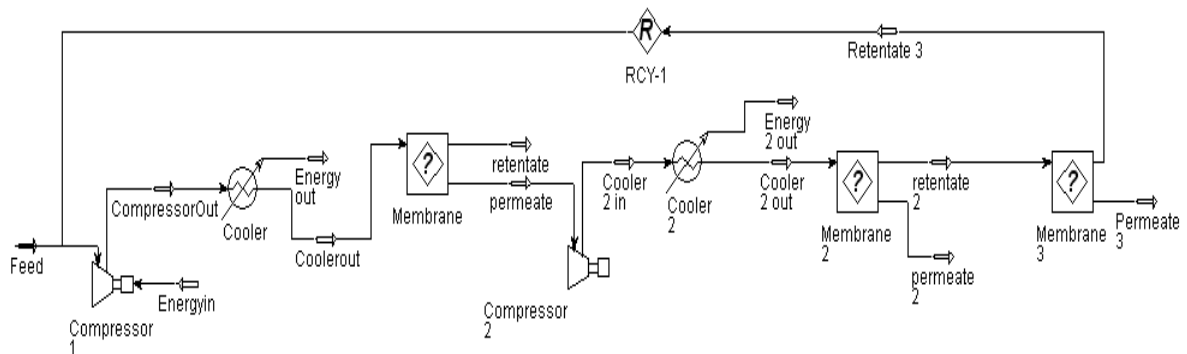


Fig. 2e. Process flow diagram (PFD) in ASPEN HYSYS for design configuration having triple stage with retentate recycle (TSRR)

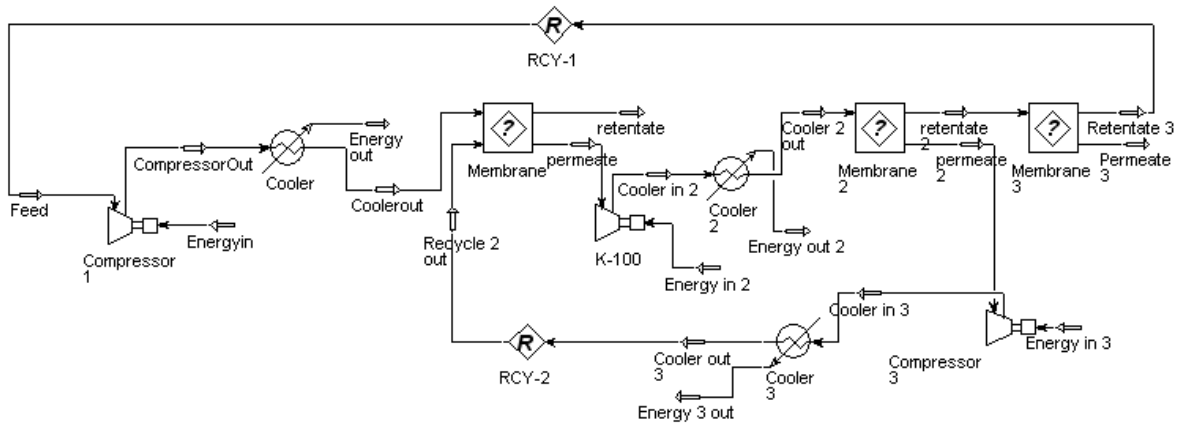


Fig. 2f. Process flow diagram (PFD) in ASPEN HYSYS for design configuration having triple stage with permeate and retentate recycle (TSPRR)

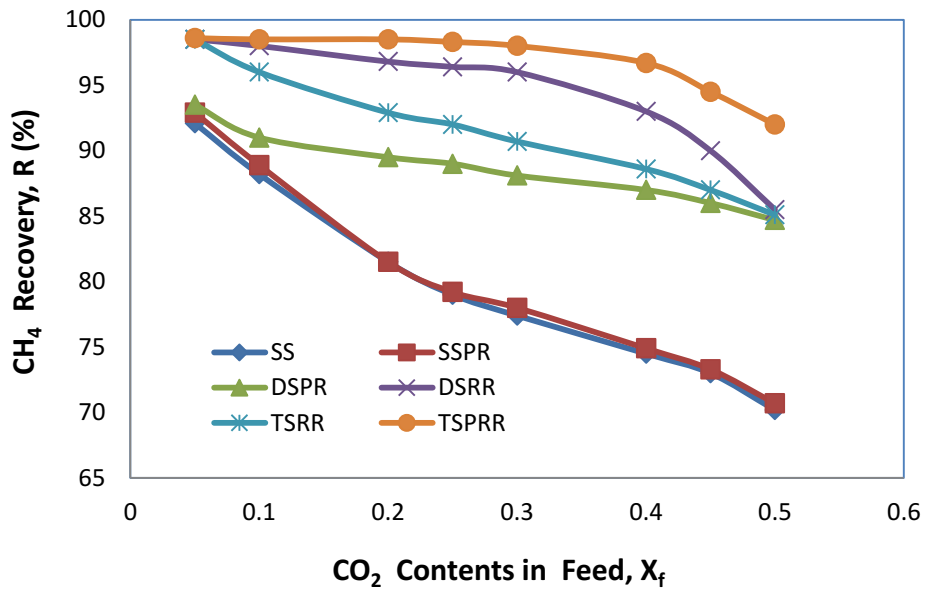


Fig. 3. Effect of feed composition on methane recovery

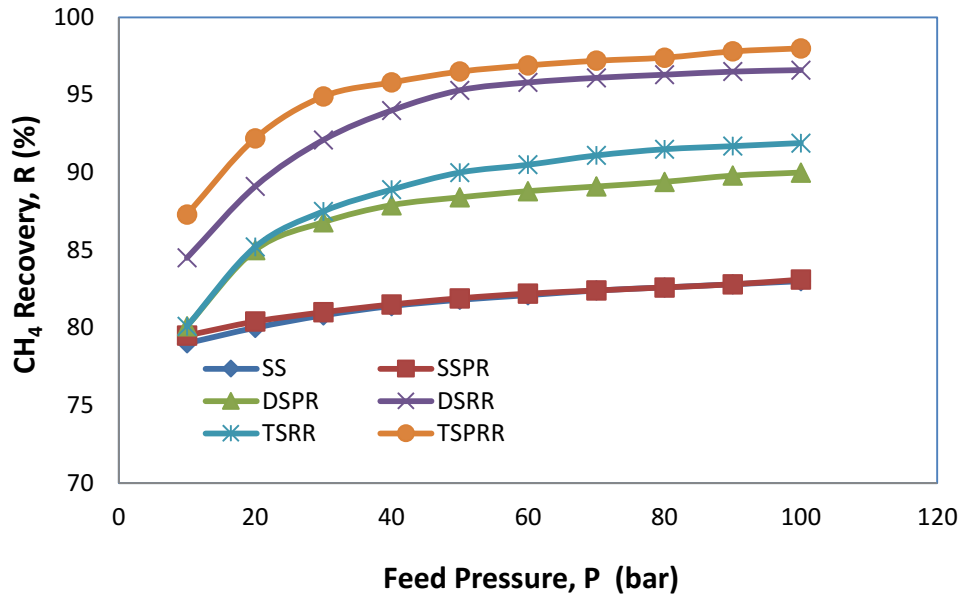


Fig. 4. Effect of feed pressure on methane recovery

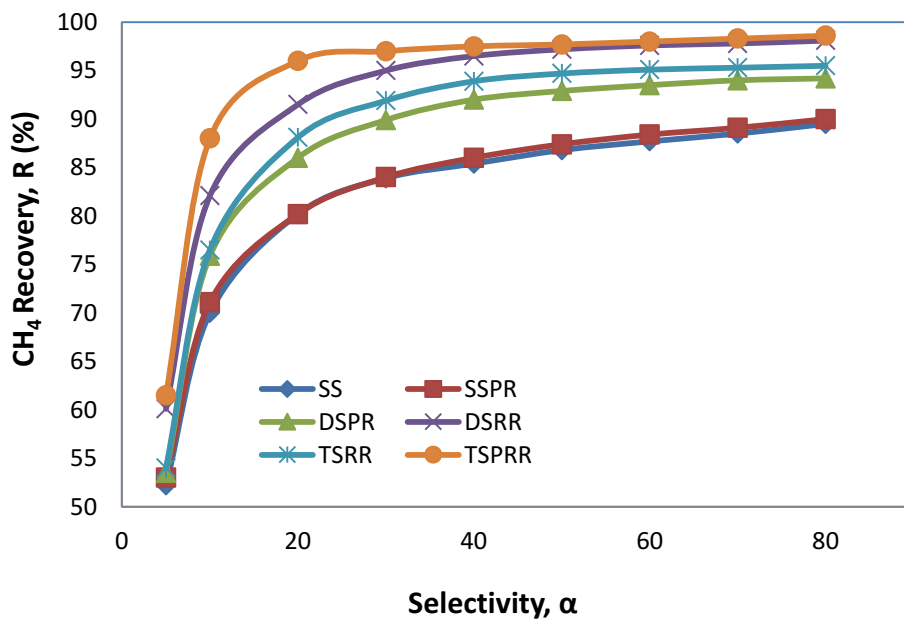


Fig. 5. Effect of membrane selectivity on methane recovery

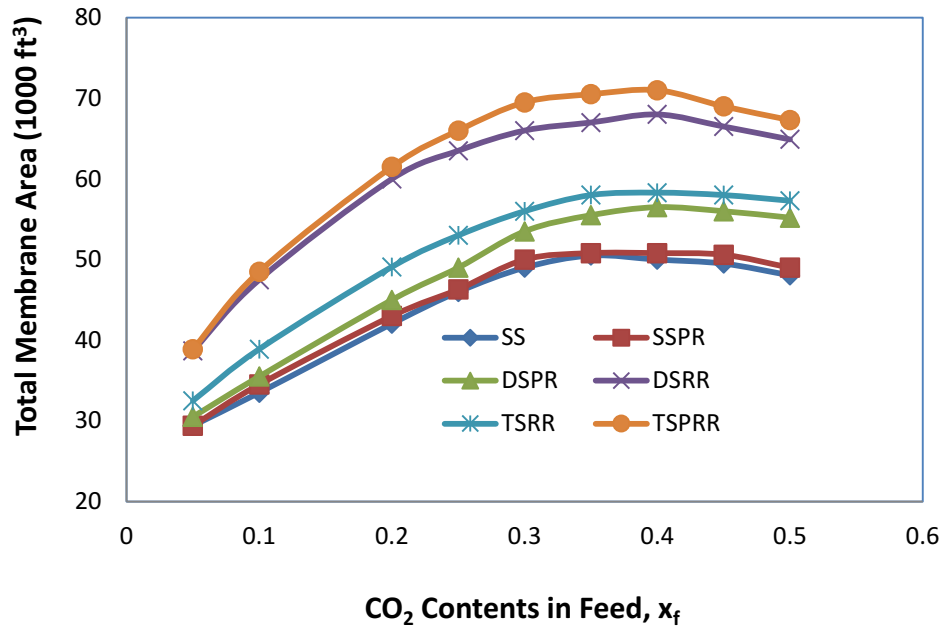


Fig. 6. Effect of feed composition on total membrane area

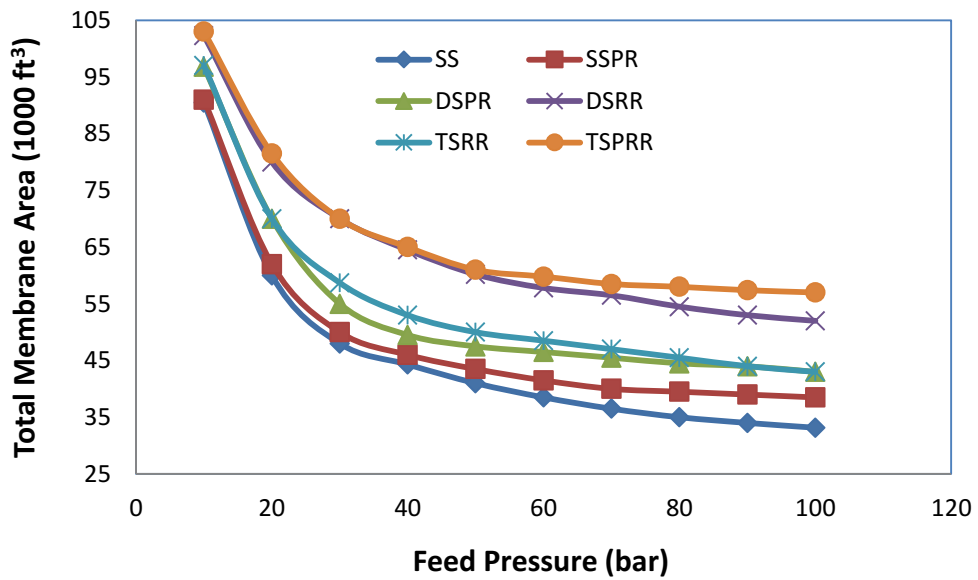


Fig. 7. Effect of feed pressure on total membrane area

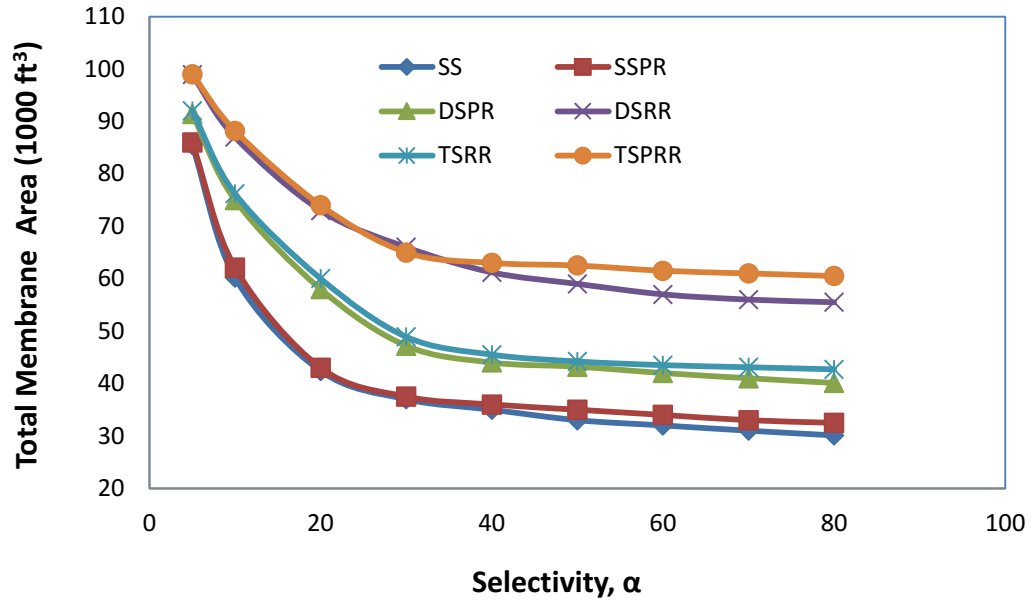


Fig. 8. Effect of membrane selectivity on total membrane area

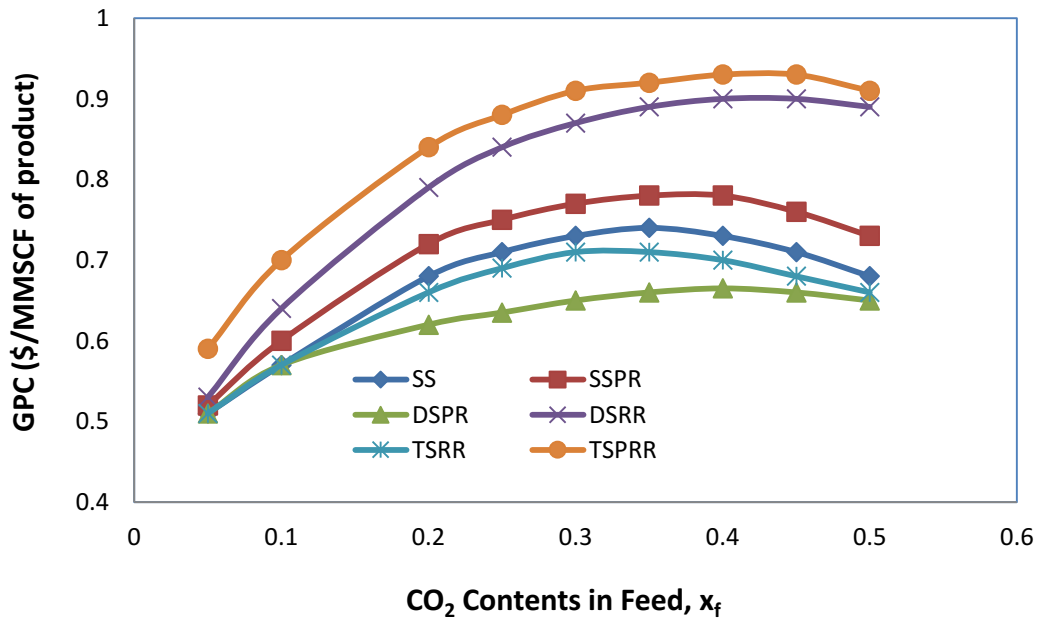


Fig. 9. Effect of feed composition on gas processing cost

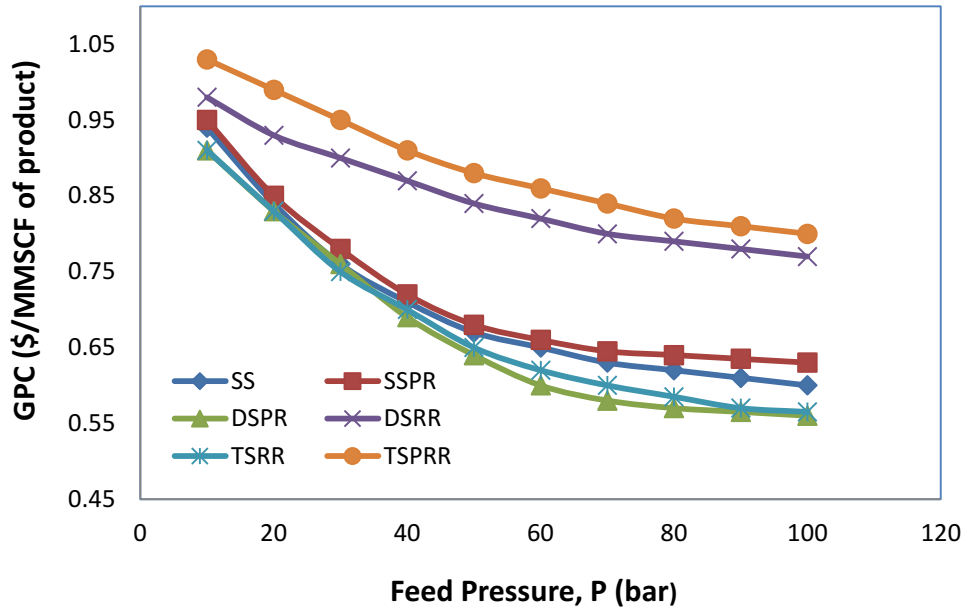


Fig. 10. Effect of feed pressure on gas processing cost

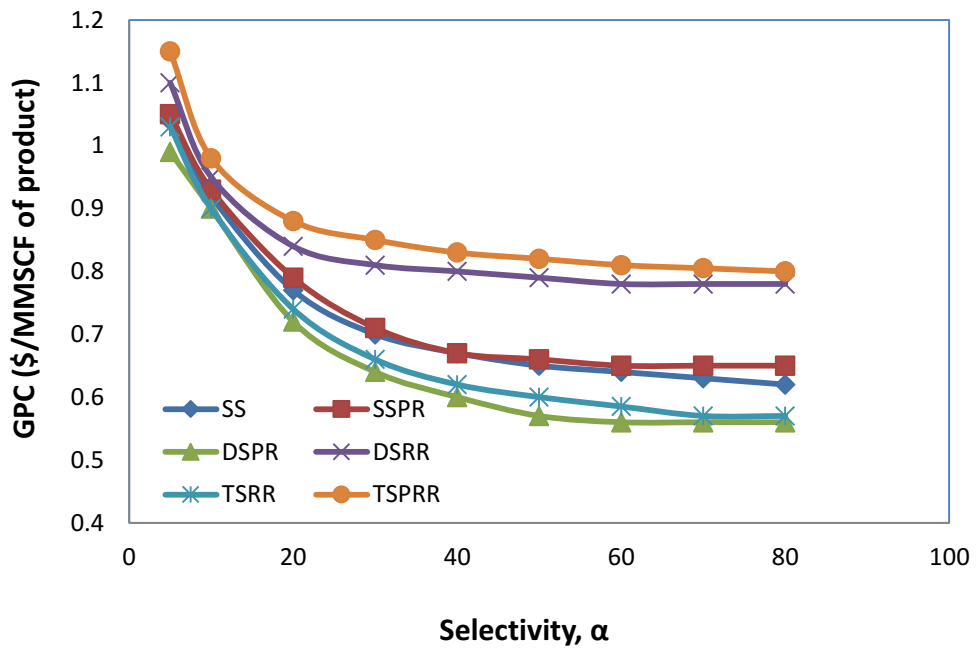


Fig. 11. Effect of membrane selectivity on gas processing cost