Investigation of Membrane Systems Efficiency in Methane Recovery or Nitrogen Removal from Waste Gas of LNG Projects by Mathematical Modeling

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ABSTRACT

Removal of nitrogen from natural gas has been important because of defects such as reduction of flame temperature, thermal value of fuel, increase in costs of transfer pipeline and pressure improving installations. Cryogenic distillation is mostly used for this purpose; however other technologies such as membrane separation and pressure swing adsorption have been suggested due to high capital investment costs. The Process and economic rationales of these separation methods depend on parameters such as contamination amount in feed, pressure and flow rate of feed and necessary purity grade of product. In this study, Investigation and design of membrane systems were done by an approximate modeling technique that proposed for spiral-wound membrane for reduction of nitrogen from the waste gas, form 33 mol. % to less than 20 mol. %, were evaluated. Designs were performed based on two types of membrane, methane or nitrogen selective membranes. The obtained results demonstrated that two-step membrane system with methane selective membrane at low flow rate (10 MMscfd) seemed more suitable from process and economical view point. Maximum recovery and purity percentages for methane were 90% and 80%, respectively but this system in high flow rate because of high capital investment costs is an uneconomical process. Generally, in high flow rate and with nitrogen content more than 15 mol. %, cryogenic distillation is the only economic process by which the obtainable purity percentage for methane could be more than 99%.

NOMENCLATURE

A membrane area (m^2)
B permeability of the spacing materials inside the spiral-wound leaf (m^2)
d thickness of membrane skin (m)
d_m thickness of membrane leaf (m)
h dimensionless leaf-length variable
L membrane leaf length (m)
P feed-side pressure (Pa)
Q_1 permeability of the more permeable component (m^3/m s Pa)
Q_2 permeability of the less permeable component (m^3/m s Pa)
R_g ideal gas constant (m^3 Pa/kg mol K)
T temperature (K)
U_f feed gas flow rate for each permeator (mol/s)
W membrane leaf width (m)
x feed-side concentration (mole fraction)
x_f feed concentration (mole fraction)
x_r residue concentration along outlet end of membrane leaf (mole fraction)
y_0 permeate concentration in bulk permeate stream at permeate outlet (mole fraction)
y' local permeate concentration on the membrane surface (mole fraction)
y'_f local permeate concentration along inlet end of membrane leaf (mole fraction)
y'_r local permeate concentration along outlet end of membrane leaf (mole fraction)
α Q_1/Q_2, membrane selectivity
γ ratio of permeate pressure to feed pressure
γ_0 ratio of permeate pressure to feed pressure at permeate outlet
μ viscosity of gas mixture (Pa.s)
θ_0 ratio of permeate flow to feed flow at permeate outlet
φ dimensionless feed-side flow rate
φ_0 dimensionless feed-side flow rate at residue outlet

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INTRODUCTION

Natural gas worldwide consumption shows a 1.8% average annual growth from 2007 to 2020 period [1]. Natural gas, a vital source of world's supply of energy is one of the cleanest and safest fossil fuel. The qualifications like new transport technologies, the remarkable reserves found, the lower overall costs and less pollution than oil and coil to cause NG as primary source in near future. Continued growth in residential, commercial and industrial natural gas consumption will increase the global natural gas consumption from 108 trillion cubic feet in 2007 to 169 trillion cubic feet in 2035 [1,2]. The high growth of natural gas consumption in recent years also has been caused unconventional hydrocarbon sources such as CoalBed Methane (CBM) and LandFill Gas (LFG) have recently drawn energy companies' attention.

NG is produced from gas field and generally contains variable amount of several contaminants such as water, light paraffins, aromatics, carbon dioxide, nitrogen, and sulphur compounds. Minor amounts of helium (less than 1 vol. %) and mercury (generally 5–300 μgNm^{-3}, in few cases more than 1000 μgNm^{-3}) can be also present [3].

For various reasons, may increase the percentage of nitrogen in the gas. For example in some regions for improvement the pressure of oil and gas wells high-pressure inert gas like nitrogen is injected that continuous injection of it causes to increase the values of nitrogen in gas flows. In addition, sometimes the gas extracted from the reservoir contains high levels of N_2 and cannot send to pipeline without treatment.

Nitrogen is inert gas and hasn't heating value and presence of it in NG although hasn't the damaging effects of corrosion on the transmission and processing facilities but due to increasing the expenses of transportation and storage (each ten percent of nitrogen in pipeline's gas increases expenses of transportation about ten percent [4]), reducing the gas heating value and environmental issues it is necessary removed from NG in the site of production.

According to the international standards, separation nitrogen from NG for concentrations more than 4-5 mol. % will be done. Typical U.S.A and Iranian natural gas pipeline specifications are shown in Table 1.

Gas containing less than 10% nitrogen can generally be used by blending it with sufficient low-nitrogen-content gas for the product to meet the pipeline specification. However, blending is no longer feasible if the gas contains more than about 10% nitrogen, so much of this gas is not currently produced. This is particularly true if the carbon dioxide and hydrogen sulfide contents are also high [7].

Table 1
Maximum value of some common compositions for natural gas delivery to the pipeline grid [5, 6]

Component	**Specification**	
	USA	IRAN
CO_2	<2%	<1%
H_2O	<120 ppm	<110 ppm (mg/Nm^3)
H_2S	<4 ppm	<5 ppm
C_{3+}	950-1050 BTU/scf; Dew point <-20 ˚C	1000-1180 BTU/scf; Dew point < -10 ˚C
N_2	<4%	<6%

One of The functional areas for such researches in Iran is recovery methane from waste gas produced in some gas projects like liquefaction of natural gas (LNG). The LNG projects are one of the appropriate methods for natural gas export. Generally, these projects have the waste gas that may include high values of methane and nitrogen that can use for power generation instead of burning in flare if reduce the nitrogen of it to acceptable value.

There are different methods for separation nitrogen from NG divided to two main categories, *cryogenic* and *non-cryogenic*.

As of 1999 to 2008 about 26 nitrogen-removal cryogenic plants were in operation in the United State [5]. Cryogenic plants are complex, require numerous moving parts and have high capital and operating costs. Furthermore, these plants must process a relatively high volume of gas before they can be run economically, typically in the range of 50 to 500 million standard cubic feet per day [8]. For these reasons, non cryogenic is considered especially when separation is in low gas flow.

Non cryogenic gas processing is growing up in recent years though it has not been able to

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completely displace other existing technologies. This process is gradually gaining market share in those applications where it has a clear economic or technical advantage. Two main technologies involved in non cryogenic process are pressure swing adsorption and membrane separation. Some advantages of Non Cryogenic Separation Processes in comparison with cryogenic separation are:

- The non cryogenic technique is much smaller in size than cryogenic plants
- The basic advantage is that it can be placed directly on the customer's site
- They can operate at near-ambient temperature and pressure
- start up and shut down of them is Easily and fast

BACKGROUND: Permeation Theory

The best measure of a membrane's ability to separate two gases i and j is the ratio of their permeabilities, $\alpha_{i/j}$; this parameter is called the membrane selectivity, and it can be written as:

$$\alpha_{i/j} = \frac{Pe_i}{Pe_j} = \frac{D_i}{D_j} \times \frac{S_i}{S_j} \quad (1)$$

The ratio D_i/D_j is the ratio of the diffusion coefficients of the two gases and can be viewed as the mobility selectivity, which indicates the relative motion of individual molecules of the two components i and j. The mobility selectivity is proportional to the ratio of the molecular size of the two permeants. In polymer materials, diffusion coefficients decrease as the molecular size increases, because large molecules interact with more segments of the polymer chain than do small molecules.

Hence, the mobility selectivity D_i/D_j always favors the permeation of nitrogen (the kinetic diameter of nitrogen is 3.64 Å and methane is 3.80 Å).

The ratio S_i/S_j is the ratio of the sorption coefficients, which indicates the relative concentration of the components i and j in the membrane material. The sorption of a component increases with condensability of the component; therefore, the sorption selectivity is proportional to the relative condensability of components i and j and methane is more condensable than nitrogen (boiling point of methane is 111 K and nitrogen is 77 K). It follows that the effects of the mobility and sorption selectivity terms for the separation of nitrogen–methane mixtures in Eq. (1) are opposed. Because the two terms are opposed, membranes can be made that selectively permeate nitrogen (maximum selectivity of nitrogen over methane ($\alpha_{N2/CH4} \approx 2.5$)), or that selectively permeate methane (maximum selectivity of methane over nitrogen ($\alpha_{CH4/N2} \approx 3$-$4$)) [5].

Current membranes used for natural gas separation applications are produced as hollow fibers or flat sheets packaged as spiral-wound modules. These modules have high ratio of area to volume and have almost lower price than tubular membrane module. Natural gas streams contain multiple components that can degrade and plasticize the membrane and also contain entrained oil mist, fine particles and hydrocarbon vapors that can easily collect on the membrane surface, In addition, the gas typically treated at relatively high pressures of 20-60 bar. Under these conditions, higher permeances of flat sheet membranes formed as spiral wound modules usually can compensate for higher cost compared to hollow fiber modules [5, 9, 10]. Also this module is used for natural gas sweetening process. Thus in this study for nitrogen removal from natural gas (methane) modeling of this module was used in process design.

MODEL INTRODUCE

The economics of membrane separation processes depend critically on the process design. A wide variety of spiral wound permeator have been proposed for both binary and multicomponent separation that differ according to assumptions about the flow pattern and permeate side pressure drop.

In 1950 and 1954 models based on the assumption of complete mixing on both sides of the membrane consist of simple nonlinear algebraic equations are proposed but such models are not sufficiently accurate for process design since complete mixing is rarely achieved in practice [11]. A muulticomponent spiral-wound model without permeate side pressure drop is proposed by Pan and Habgood in 1978 [12] and also models based on plug flow and cross flow patterns are proposed by Pan (1983), Chern and et al. (1985), Li and et al. (1990) that while such models offer improved accuracy as compared to complete mixing models, they are likely to result in prohibitive computational requirements when utilized for process design [11]. An alternative approach is to developed approximate permeators models that offer a better tradeoff between prediction accuracy

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and computational complexity. Qi and Henson in 1996 proposed an approximate model technique for spiral wound permeators separating binary gas mixtures that in this study we used this model for N_2/CH_4 separation evaluation by spiral wound membranes. The model development is based on figure 1 and is derived from the fundamental model that is based on mass balance.

The basic assumption considered in the proposed model is the residue flow rate does not vary in the direction of permeate flow. Other assumptions are on the following:

1. The feed stream contains a binary gas mixture at a relatively high pressure.
2. Permeation is described by a cross flow pattern.
3. The pressure drop on the permeate side is described by the Hagen-Poiseulle equation.
4. There is no pressure drop on the feed side and equal to the pressure of the feed stream.
5. Membrane permeability is independent of pressure and concentration.
6. The permeate pressure varies only in the direction of permeate flow.

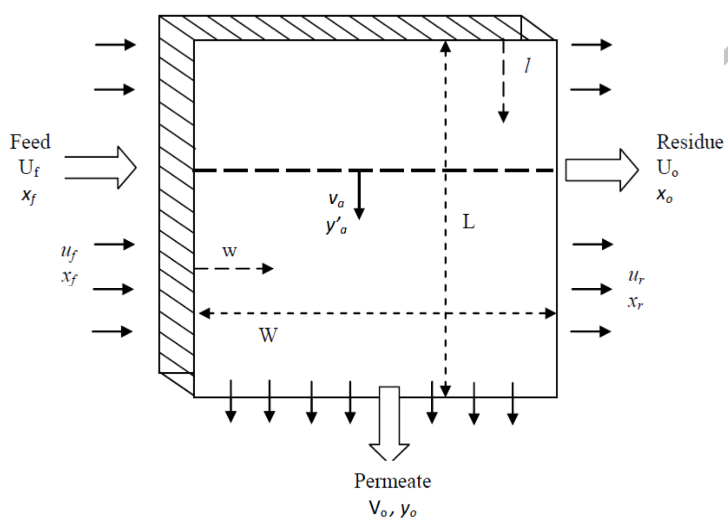

Figure 1
Gas permeation through a spiral-wound membrane [11]

The approximate model is comprised of four groups of nonlinear algebraic equations. The first group describes the permeate side pressure distribution,

$$\gamma^2 = \gamma_o^2 + \frac{1}{2} C (1-\varphi_r)(1-h^2) \quad (2)$$

Where γ(h) is the ratio of the permeate side pressure to feed side pressure and γ_0 is γ at the permeate outlet. $\varphi_r(h)$ is the dimensionless residue gas flow rate; h is the dimensionless membrane leaf length variable and:

$$C = \frac{2R_g T \mu L U_f}{W d_m B P^2} \quad (3)$$

The remaining variables are defined in the legend and it is necessary to mention that, where the meaning of concentration of gas is mole fraction.

The coefficient C can be factored as follows:

$$C = C'' \frac{U_f}{AP^2} \quad (4)$$

Where C'' is a parameter that depends on the internal properties of the permeator.

The second group describes the effect of γ and the local permeate side concentration, y', on the dimensionless feed side flow rate, φ,

$$\varphi(\gamma, y') = \frac{u}{u_f}(\gamma, y') = \left(\frac{y'}{y'_f}\right)^a \left(\frac{1-y'}{1-y'_f}\right)^b \left(\frac{\alpha-(\alpha-1)y'}{\alpha-(\alpha-1)y'_f}\right) \quad (5)$$

Where a and b are:

$$a = \frac{\gamma(\alpha-1)+1}{(\alpha-1)(1-\gamma)} \qquad b = \frac{\gamma(\alpha-1)-\alpha}{(\alpha-1)(1-\gamma)} \quad (6)$$

The dimensionless feed side flow rate at the residue outlet can be written as,

$$\varphi_r = \frac{u_r}{u_f} = \varphi(\gamma, y'_r) \quad (7)$$

The third group describes the relation between the dimensionless permeation factor, R,

$$R = \frac{2WLQ_2}{dU_f} = A \frac{Q_2}{d} \frac{P}{U_f} \quad (8)$$

and the local permeate concentration along the residue outlet, y'_r :

$$R = \frac{1}{\alpha(1-\gamma)} \{\alpha - (\alpha-1)y'_f - (\alpha-(\alpha-1)y'_r)\varphi(\gamma, y'_r) - (\alpha-1) I(\gamma, y'_r)\} \quad (9)$$

The subscript "2" represents base component in membrane selectivity ($\alpha = Q_1/Q_2$).

Here $I(\gamma, y'_r)$ is an integral function which is approximated using Gauss-Legendre quadrature.

The integral is represented as:

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$$I(\gamma, y'_r) = \int_{y'_f}^{y'_r} (\frac{U}{U_f})_\gamma dy' = \int_{y'_f}^{y'_r} \varphi_\gamma(y')dy' \approx \frac{y'_r - y'_f}{2}\sum_{j=0}^{n} w_j \varphi_\gamma(\frac{z_j(y'_r - y'_f) + (y'_r + y'_f)}{2}) \quad (10)$$

In eq. (10), w_j is weight function and z_j is roots of Legendre polynomial that used in this method of integration.

The fourth equation describes the relation between the feed side concentration, x, and the local permeate side concentration, y':

$$\frac{y'}{1-y'} = \frac{\alpha(x-\gamma y')}{1-x-\gamma(1-y')} \quad (11)$$

The residue concentration $x_r(h_i)$ is obtained from Eq. 11 whit $y'=y'_r(h_i)$

The flow rate and concentration of effluent permeate stream are calculated by integration along the length of membrane that these integral expressions also are approximated using the Gauss-Legendre quadrature:

$$\theta_0 = 1 - \int_0^1 \frac{u_r}{u_f} dh \approx 1 - \frac{1}{2}\sum_{i=0}^{m} w_i \times \varphi_r(\tfrac{1}{2} z_i + \tfrac{1}{2}) \quad (12)$$

$$y_0 = \int_0^1 y'_a dh \approx \frac{1}{2}\sum_{i=0}^{m} w_i \times y'_a(\tfrac{1}{2} z_i + \tfrac{1}{2}) \quad (13)$$

The results have showed that under most condition, solution of integral statements with a single point quadrature usually offers satisfactory solution [11, 12, 13]. So in this state $z_i=0$ and $w_i=2$ and the resulting equations (12), (13) are:

$$\theta_o = 1 - \frac{u_r}{u_f}(h_i) \quad (14)$$

$$y_o = y'_a(h_i) = \frac{x_f - x_r(h_i)\frac{u_r}{u_f}(h_i)}{1 - \frac{u_r}{u_f}(h_i)} \quad (15)$$

It is interesting to note that the pressure distribution (2) has the following form in this case:

$$\gamma^2(h_i) = \gamma_o^2 + \frac{3}{8}C[1 - \frac{u_r}{u_f}(h_i)] \quad (16)$$

Simultaneous solution of the equations (2), (7), (9), (11) with $x=x_f$ at the each point of h_i yields $\gamma(h_i), \varphi_r(h_i), y'_r(h_i)$ and $y'_f(h_i)$. Note that h_i is value of h in different points of quadrature (it means ($h_i = \frac{1}{2} z_i + \frac{1}{2}$)).

The flow rate and concentration of the residue stream are determined from an overall material balance about the permeator.

$$\eta_o = 1 - \theta_o \quad (17)$$

$$x_o = \frac{x_f - \theta_o y_o}{1 - \theta_o} \quad (18)$$

Furthermore, in this study calculations were done base on single point formula (M=0) for equations (12), (13) and three points formula (N=2) for equation (10). Note that three points formula for polynomial functions with order 2n+1=5 or less gives acceptable answer and maximum order of y' in equation (5) is frequently less than this value.

MODEL PARAMETERS

In most applications, detailed characteristics of the permeator are not known at the preliminary design stage. Consequently, the approximate model may contain uncertain or unknown parameters. Unknown parameters may include the membrane leaf length, L, the leaf width, W, the thickness of membrane skin, d_m, and thickness of membrane leaf, d, that these four parameters appear in the approximate model only via the terms C and R. Because the parameters C and R depend on feed flow rate and feed pressure, so we can estimate C'' and Q_2/d that don't vary with the operating conditions. Also in this work, we allow the membrane area to be a variable.

Where, Q_2/d and α have been selected from Richard W. Baker and et al. [10] that are shown in table 2, and C'' was estimated from the simulation results that reported in it.

Table 2
Membrane performance used for process design simulations [10]

Gas	Methane selective membrane		Nitrogen selective membrane	
	Permeances (gpu*)	Selectivity Gas/Nitrogen	Permeances	Selectivity Gas/Methane
Nitrogen	50	1	50	2.5
Methane	150	3	20	1
Permeance (Q_2/d), ($m^3/m^2.s.Pa$)	3.8×10^{-10}		1.52×10^{-10}	

*gpu= gas permeation unit= $10^{-6} cm^3(STP)/cm^2.s.cm\ Hg$

PROCESS DESIGN AND ECONOMICS

The complexity and cost of the methane/nitrogen membrane separation process increases with the

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nitrogen content of the gas, especially if the product gas from the separation is targeted to contain low-nitrogen content. In the process design, the first object is reduction of nitrogen to specified amount and also minimizes methane loss simultaneously.

Single stage membrane units are rarely used for this separation but they do provide a good example for the difference between the two types of membranes.

The design of the process depends on the concentration and the pressure of the gas to be treated and the specification for the product gas [14].

In natural gas treated by membrane more commonly, a two-step or two-stage membrane system is required. The system contains a second membrane step when the second membrane unit is placed on the residue gas from the first membrane unit and the system contains a second membrane stage when the second membrane unit is placed on the permeate gas from the first membrane unit.

In this study, the desired waste gas is produced in LNG project at Tonbak in Iran that specifications of it have been exerted in table 3. Due to heating value of this gas and also environmental constraints, it is recommended that this gas is consumed as fuel in gas turbines used in LNG process. The gas for use in gas turbine must have minimum required quality that some of them have been shown in table 4.

Table 3
The Specifications of available Fuel gas [15]

Pressure(bar g)	32
Temperature(˚C)	44
Heating Value(kJ/kg)	27270
Percentage of Component molar fraction	
Water	0.069
N_2	32.529
CO_2	0
H_2S	0
Methane	66.703
Ethane	0.419
C_{3+}	0.349

Table 4

The properties and conditions of the fuel gas that must be satisfied at the terminal point of supply of the gas turbine supplier [15]

Temperature	50 ˚C (max.) & -10 ˚C (Min.)
Pressure	21 bar (abs.)
CH_4	≥ 80 mol.%
Ar , N_2 ,CO_2	≤ 20 mol.%

Thus the proposed system must reduce nitrogen less than 20 mole percent.

RESULT AND DISCUSSION

A number of design studies were performed in the figures 2-7, to determine the best process configurations. These calculations were performed for both methane-selective and nitrogen-selective membranes, using the model that introduced in previous sections. The membrane properties used in the process design calculations are shown in Table 2. In these conceptual designs, we assume the gas to be treated consists of 33% nitrogen and 67% methane only, all membrane units operate at 30 ˚C, and the feed gas flow is 10 MMscfd of gas at 32 barg and ratio of permeate pressure to feed pressure at outlet membrane (γ_o) is 0.1. In these calculations, the sizes of the membrane units were adjusted so that the recycle stream mixing with the feed gas has the same nitrogen concentration as the feed gas.

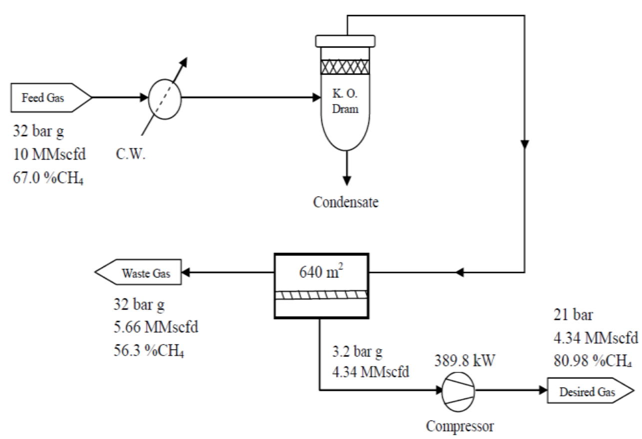

Figure 2
Single stage system by Methane selective membrane

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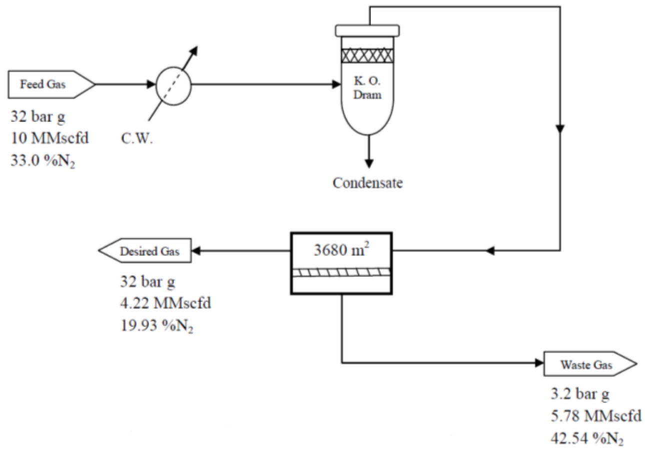

Figure 3
Single stage system by Nitrogen selective membrane

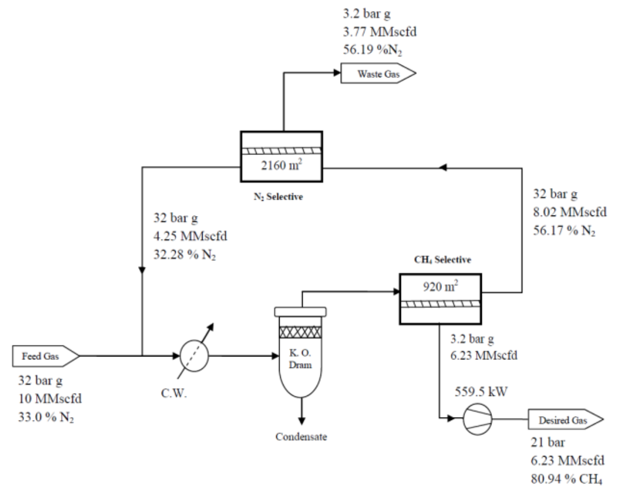

Figure 6
Tow step system by Methane & Nitrogen selective membranes

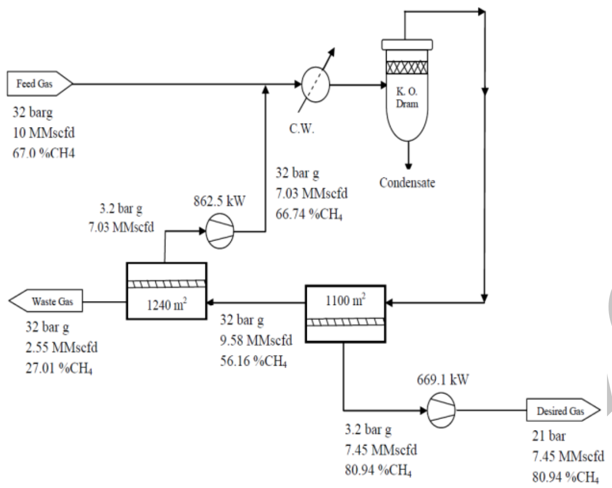

Figure 4
Tow step system by Methane selective membrane

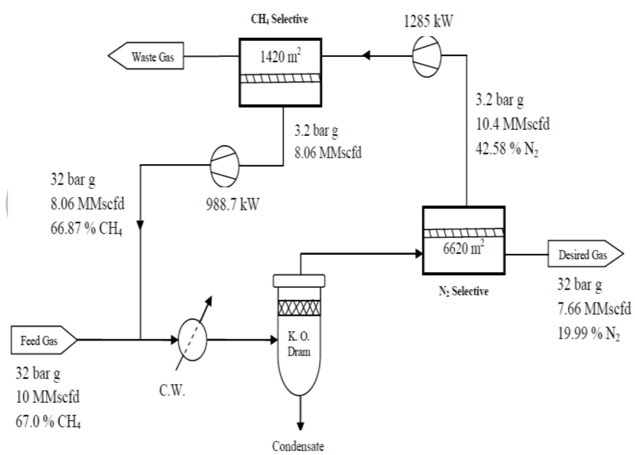

Figure 7
Tow stage system by Methane & Nitrogen selective membranes

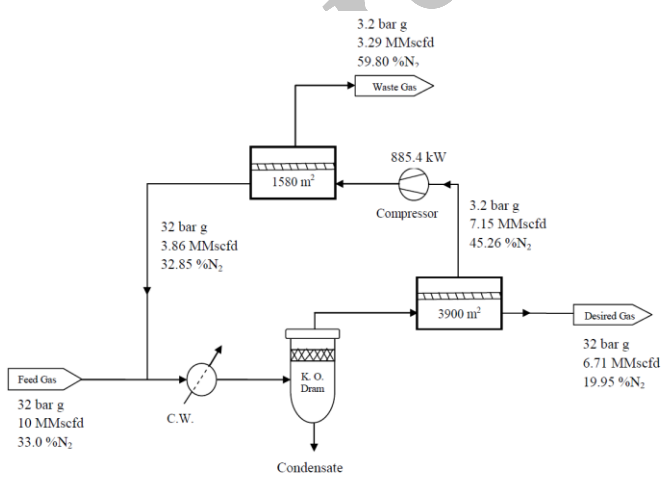

Figure 5
Tow stage system by Nitrogen selective membrane

Table 5 provides a cost summary for separation of nitrogen from natural gas for each process shown in Figures 2 to 7.In cost estimation, the base prices obtained from the references [14], [16] and then Marshall and swift cost index was applied to calculate the equivalent price in 2010.

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Table 5
Comparison of Capital Costs of the Six Nitrogen Gas Separation Systems Illustrated in Figures 2 to 7

Operating Conditions and Cost Elements	**Fig. 2**	**Fig. 3**	**Fig. 4**	**Fig. 5**	**Fig. 6**	**Fig. 7**
Membrane area (m^2)	640	3680	2340	5480	3080	8040
Compressor Power (kW)	389.8	-	1531.6	885.4	559.5	2273.7
Methane recovery (%)	52.5	49.6	90.0	80.2	75.3	91.5
Capital Cost (1000 $)						
Membranes at $250/$m^2$	160	920	585	1370	770	2010
Module housings (5×20m^2 modules/housings at $5,300)	34	49	31	73	41	107
Purchased cost of Compressor (centrifugal)	330	-	1056	660	460	1200
Total	524	969	1672	2103	1271	3317
Product gas (MMscfd)	4.34	4.22	7.45	6.71	6.23	7.66
Capital cost (1000$)/ MMscfd of product gas	120.74	229.6	224.4	313.4	204.0	433.0

CONCLUSIONS

In nitrogen/methane separation, methane selective membrane has been usually used. This approach, requires recompression of permeate flow that may affect on the economic calculations of process. Here, the cost calculations show that use of one stage system by methane selective membrane has better result compared to nitrogen selective membrane.

Among of all proposed systems, figure (7) has the most of methane recovery but the capital cost of it is very more than other cases. Finally, figure (4) was diagnosed as the best in terms of price and methane recovery.

The economics of membrane process are affected by the nitrogen content in the feed gas, the value of the product gas, and the maximum allowable nitrogen content in the product gas. For example above systems have been evaluated for a gas flow of 10 MMscfd and in higher flow rate may be non-economic system. Generally, in high flow rate and with nitrogen content more than 15 mol. %, cryogenic distillation is the only economic process by which the obtainable purity percentage for methane could be more than 99%.

KEYWORDS
Nitrogen removal, Membrane permeation, Membrane Modeling, nitrogen/methane separation, spiral wound membrane, process design, natural gas, pipeline specification

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