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MULTI-OBJECTIVE OPTIMIZATION OF A COMBINED CRYOGENIC AND MEMBRANE PROCESS FOR HELIUM RECOVERY FROM NATURAL GAS USING GENETIC ALGORITHM

Mohammad Heidari[1,*], Ahmad Tavasoli[1,2*], Ali Karimi[2]

[1]Department of Energy Engineering, Graduate School of the Environment and Energy, Science and Research Branch, Islamic Azad University, Tehran, Iran; [2]School of chemistry, college of science, University of Tehran, Tehran, Iran

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Abstract

Increasing demand for Helium and also high energy requirement of Helium production make the necessity of improving Helium recovery processes. This study is aimed to simulate and optimize a combined cryogenic and membrane process for He separation from natural gas. APCI flashing based system and two stage membrane system are chosen for cryogenic and membrane sections respectively. The proposed combined system can produce a 99% containing Helium gas stream. In order to enhance the performance of the system, a parametric study and exergy analysis are conducted to determine the effects of the feed gas pressure on the exergy efficiency. Moreover, a complete optimization study is carried out by a multi-objective evolutionary based genetic algorithm (NSGA-II) considering two different objective functions, exergy efficiency and total cost rate. The results indicated that when considering both objective functions with equal importance, 42.47% and 0.06185 \$/s were obtained at the optimal point for exergy efficiency and total cost rate respectively. The feed gas pressure at this point was 2006 kPa, the inlet feed pressure to the membrane was 1486 kPa and the required membrane area was 1710 m^2.

Keywords: Cryogenic; Membrane; Helium; Exergy; Economic; Genetic algorithm.

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1. Introduction

Helium is a precious element and it is widely used in industry such as cryogenics and welding due to its unique properties. However Helium market is becoming tighter as production is not keeping up with demand. Moreover, Helium supply is easily affected when one of any Helium production plants in the world is become offline, as there are only few [1]. This situation indicates the importance of discovering new methods or new sources for Helium production. The main source of this rare element is natural gas and extracting it from LNG end-flash has become one of the major production approaches. Helium has lower boiling point of -269°C. Other constituent of natural gas such as Methane has boiling point of about -161°C and nitrogen has boiling point of about -196°C. Certain processes for using and shipping natural gas, such as the production of liquefied natural gas (LNG), may generate the temperatures needed to isolate Helium. This is done by lowering the gas temperature until almost all other components are liquefied, except the Helium [2]. The reliance on cryogenic methods means that the recovery of He from a gas reserve is not just dependent on the He content of the reserve but also strongly dependent on development of LNG production facilities. Although, the cost of He extraction may be relatively small compared to the total LNG plant costs, the additional capital cost of cryogenic He extraction and purification plants are significant and investment decision to install them is still required [3].

One of the major cryogenic approaches is flashing based processes. In the multi-stage flash process, the pressure of the LNG feed, which contains the dissolved Helium and some dissolved Nitrogen, is reduced in a series of flash vessels. In each flash stage, the He will be vaporized along with the N_2. The liquid NG product is used to make the feed colder. Several flashing based processes have been cited in literature. As instance, Roberts et al. invented a flashing based He extraction process, claiming to recover He from feed streams to an LNG plant that has less than.1 vol% He in the feed [4]. Bauer et al. also developed a multi-stage flash cycle process for He production in a German patent [5]. They used a different arrangement than the previous works which helped to produce more and richer crude He. Moreover, He could be purified by no cryogenic techniques such as membrane separation. The membrane separation process turns out to be a relatively slow and low-pressure process [6]. Existing studies that most commonly employ membrane technologies, involve mixtures of hydrogen with gases other than He [7]. He extraction plants using membrane separation have reportedly been used for a long time. Laverty and O'Hair at 1990 report a membrane process for He recovery in Alberta, Canada [8]. However, not much data on the performance or operating experiences of these membrane-based He recovery plants have been published in the open literature. By contrast, the use of H_2-permeable membranes in the production and purification of H_2 has been described in the literature. Because H_2 and He are so similar in size, most membranes have very close permeability for these gases. Successful systems for separating hydrogen from methane or carbon dioxide at high temperatures (450-900°C) in La_2NiO_4-zeolite membrane reactors are reported by Liu *et al.* at 2001 [9] and Jordal *et al.* at 2004 [10].Most membrane processes cannot achieve simultaneously a high product purity and high product yield (throughput). To increase the purity of the product (i.e. He), several membrane stages can be operated in series, However, due to the large pressure drop of the He-rich permeate across the membrane unit, multistage membrane processes require inter-stage compressors. The recompression between membrane stages increases both capital and operating costs of membrane systems [3]. Other similar studies could be mentioned in He membrane. As instance, Hale et al. described membrane systems for the recovery of He from reject streams in gas processing, which may include vent streams, fuel gas streams and purge streams and claimed that the process may use any type of membrane that can provide an adequate He flux and a high selectivity for He over the other principle components of the reject gas (N_2, CH_4, CO_2) [11]. Baker *et al.* [12] also explained a process for the removal of N_2 from NG using two or more membrane stages. An example including the concentration of He in the N_2-rich product is also described in their patent. The process uses a CH_4-selective membrane operating at -40 to 0°C in the first stage followed by an N_2-selective membrane in the second stage [12]. Methods well known in the field of high-purity He gas production may include a combination of the cryogenic separation process and a gas separation membrane process. As instance, a method of producing high-purity Helium is described in Shoji et al. patent. The inventors of this invention have found that, to obtain high-purity He from crude He having a He purity of about 40 to about 90mol %, which is obtained by an arbitrary process with a cryogenic separation unit and the like, with natural gas as the feed Hewith high purity of 99.99mol % or higher can be obtained by using a glass hollow fiber membrane module. According to the this invention, because He and nitrogen can be separated from each other with high selectivity, a refrigerating power necessary to cool to -200C is needed to liquefy and separate the nitrogen with a conventional cryogenic separation unit for producing high-purity He gas, becomes unnecessary [13].

Although, there could be find few combined cryogenic and membrane processes in the patents but no study have evaluated and optimized them simultaneously from different aspects. In this study, first Helium recovery from a natural gas by a cryogenic flashing process is simulated. Through this step a mixture with about 60% He could be achieved. Afterwards a two stage membrane process would be used to obtain a 99% Helium stream. Parametric study, exergy, economic and environmental analysis are performed to investigate the effects of the feed pressure on the hybrid membrane-cryogenic system performance. An EES coding which is popular in thermodynamic affairs is developed for thermodynamic modeling of the system.

Afterwards, the results of the EES coding are used to drive objective functions by polynomial regression in Excel and finally multi-objective optimization considering exergy efficiency and total cost rate as objective functions is carried out by Non-dominated Genetic algorithm in MATLAB.

2. Material and methods

Natural gas composition obtained from South Pars gas field is reported in Table 1. The flow rate of this gas which consists 0.04% He is 2.77 kg/s. Helium content in the feed gas is one of the major factors that influences the performance and economics of a helium extraction facility. The feed gas was assumed as dehydrated, sweetened natural gas, containing low amounts of heavy hydrocarbons.

Table 1 Feed gas composition

Component	Fraction on total gas	Component	Fraction on total gas
Helium	0.0004	i-Butane	0.0011
n-Hexane	0.0001	Propane	0.0053
n-Pentane	0.0001	Ethane	0.0160
i-Pentane	0.0002	Methane	0.9541
n-Butane	0.0018	Nitrogen	0.0209
Pressure		1120 kPa	
Temperature		35^{o}C	

2.1 System description

A hybrid cryogenic-membrane system is simulated to achieve a 99% Helium content stream. However, LNG and fuel streams are also obtained from the cryogenic section. The cryogenic flashing based process is originally invented by APCI, the world largest Helium producer [14]. This process is designed to recover Helium from feed streams that has less than 0.1 % He in the feed, which is appropriate for LNG plants. Since there have been no technical papers about this system and no detailed data to know about the process conditions, it is modeled in HYSYS version 8 first and some design parameters are assumed reasonably. A schematic of the simulation is illustrated in Figure 1. As shown, first, treated feed gas (1)which may be compressed and then cooled (it is related to optimization part and not shown in this figure), splits into stream 2 and 24. Stream 2, is sent to an LNG liquefier and so sub-cooled LNG (3) is obtained. Stream 24containing a lower portion of the feed is sent to a heat exchanger for cold recovery. Next, 24 is condensed and sub-cooled to the temperature of3 in the heat exchanger. Then, 3 and 25, are mixed to make 4 and it is throttled by the first throttling valve to produce 6 and 7 by a flash tank (FT1).This is repeated in Flash Tank-2 and Flash Tank-3 and each time the bottom stream of the flash tank is more Helium stripped. Finally, the LNG stream (12) is taken from the bottom of the third flash tank (FT3).

Stream 7 is cooled significantly and partially condensed by heat exchanger (Exchanger1) to go through a phase separator (FT4) for generating15,a He rich gas from top of the separator. The bottom stream of the phase separator is split to 17 and 18 which are depressurized to different levels to go through the heat exchanger for producing cold duty and then combined with the second and third flash tank top streams, respectively.

For recovering the remaining coldness of the first heat exchanger (Exchanger1) streams, they are delivered to another heat exchanger (Exchanger2). The outputs of the second heat exchanger go through some pressurizing and temperature controlling steps to meet the desired specifications of final fuel stream (35) and final Helium rich stream from the cryogenic section (36). Stream 36 containing approximately 60% Helium is sent to a membrane process. The schematic of this process is depicted in Figure 2.

After the first membrane, the permeat stream (38) is sent to the second membrane for further purification. It is worthy to note that before the second stage of membrane process, the stream is pressurized and then cooled to meet the desired working pressure and temperature of the membrane.

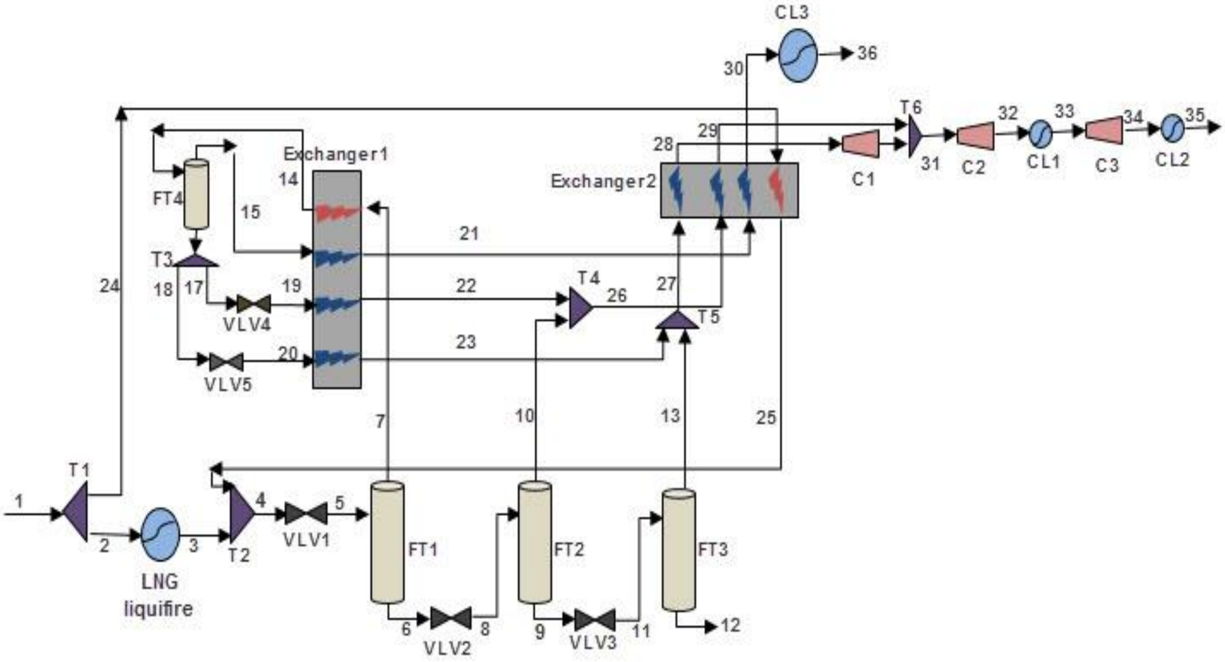

Figure 1 Schematic of the cryogenic flashing process

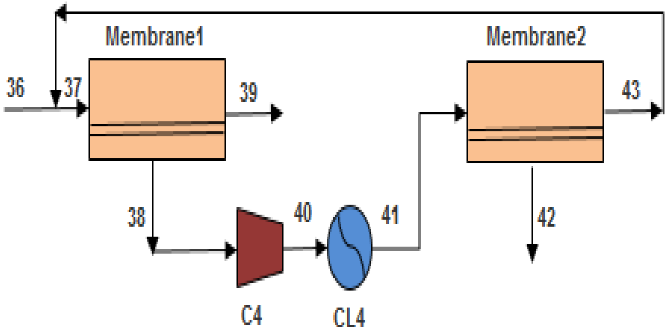

Figure 2 Schematic of the membrane process

2.2 Thermodynamic analysis

For realizing the governing equations and parametric study of the hybrid system, a thermodynamic model is developed. In the model, each component of the system is treated as a control volume and the principle of mass conservation and the first and second laws of thermodynamics are applied. The performances of the system are investigated by solving the corresponding equations together with the thermodynamic relations through an EES coding. Applying the first law of thermodynamics, energy rate balance for each component of the system under steady state condition can be determined by:

$$\sum(\dot{m}h)_e-\sum(\dot{m}h)_i=\dot{Q}_{cv}-\dot{W}_{cv} \quad (1)$$

Subscripts i and e denote inlet and outlet of control volume. By applying the second law of thermodynamics the following equation is obtained for exergy balance:

$$\sum_i \dot{m}_i ex_i + \dot{E}_Q = \sum_e \dot{m}_e ex_e + \dot{E}_W + \dot{E}_D \qquad (2)$$

E_D is the exergy destruction rate, E_Q, indicates exergy of heat transfer in a system and E_w is equal to the work transferred in the system.

$$\dot{E}_Q = (1-\frac{T_0}{T_i})\dot{Q}_i \qquad (3)$$

$$\dot{E}_W = \dot{W} \qquad (4)$$

In equation (2) term ex is the total exergy flow which consists of four components that are physical, kinetic, potential and chemical exergy. Since the changes in speed and elevation are negligible in this study, Effects of potential exergy and kinetic exergy are not considered in analysis of components of the systems. Chemical exergy is associated with the departure of the chemical composition of a system from that of the environment [15], and the physical exergy is the maximum work which can be achieved from a substance which passes to dead state from its original state through a fully reversible process which exchange heat only with environment and is defined by:

$$\dot{E}_{Ph} = \dot{m}\,[(h-h_0)-T_0(S-S_0)] \qquad (5)$$

An approach to calculate the exergy efficiency of the whole system is to calculate the exergy destructions in all of the equipment, then identifying all of the exergy supplied of the system and using the below equation:

$$\eta_{\text{Second law}} = 1-\frac{\sum \dot{E}_D}{\sum \dot{E}_{in}} \qquad (6)$$

In addition, stream-wise and source-sink approaches are recommended for exergy analysis. Stream-wise method is based on the exergy balance between input and output streams of a component. (Equation (2)). One of the major problems of stream-wise method is its low accuracy whenever the inlet and outlet conditions are close to each other [16].Basis of source-sink method is assuming a source and a sink of exergy for each operational unit of the system. According to this method, in each exergy transfer from the source to the sink, a part of exergy is lost ($\dot{E}_D$) which is given by [16]:

$$\dot{E}_D = \Delta ex_{source} - \Delta ex_{sink} \qquad (7)$$

Based on the above assumptions and principles, the equations for calculating exergy destruction in each component will be as bellow:

a) LNG liquefier: LNG liquefier is a heat exchanger. Exergy destruction can be achieved by calculating exergy changes in hot and cold streams which are source and sink of exergy respectively:

$$\dot{E}_{D,liq} = \Delta H(1-\frac{T_0}{\Delta T_{LM(T_{h,in}\&T_{h,out})}}) - \Delta H(1-\frac{T_0}{\Delta T_{LM(T_{c,in}\&T_{c,out})}}) \qquad (8)$$

b) Expansion valves: Expansion valve process is isenthalpic. The inlet is a high pressure liquid and the outlet is a two phase fluid. It is assumed that this process consists of two steps. Through the first step, temperature of inlet fluid decreases in a constant pressure (source) and through the second one the given exergy from the first step is used to evaporate some moles of the fluid in a constant temperature (Two phase flow).

$$\dot{E}_{D,liq} = \Delta H\left(1-\frac{T_0}{\Delta T_{LM(source)}}\right)-\Delta H\left(1-\frac{T_0}{T_{2nd\_step}}\right) \qquad (9)$$

c) Flash tanks: The process of flash tanks is isenthalpic, but due to composition changes of outlet flows and inlet flow, a change in exergy occurs. Change in exergy of composition for each stream and exergy destruction of a flash tank can be obtained from equations (10) and (11) respectively. Where y_i and n_i are the mole fraction and number of moles of component i

$$\Delta Ex_{OC} = RT_0\sum_{i}^{n} n_i \ln\frac{1}{y_i} \qquad (10)$$

$$\dot{E}_{D,FT} = \Delta Ex_{oc}(feed)-[\Delta Ex_{oc}(top)+\Delta Ex_{oc}(bottom)] \qquad (11)$$

d) Mixers, heat exchangers, coolers and compressors: From stream-wise approach, exergy destruction of mentioned equipment can be determined from equations (12) to (15).

$$\dot{E}_{D,Mixer} = \sum \dot{E}_{in} - \dot{E}_{out} \qquad (12)$$

$$\dot{E}_{D,Exchanger} = \sum \dot{E}_{in} - \sum \dot{E}_{out} \qquad (13)$$

$$\dot{E}_{D,CL} = \dot{E}_{in} - \dot{E}_{out} + \dot{E}_{Q} \qquad (14)$$

$$\dot{E}_{D,C} = \dot{E}_{in} - \dot{E}_{out} + \dot{W}_{C} \qquad (15)$$

$\dot{E}_{in}$ and $\dot{E}_{out}$ are the physical exergies of inlet and outlet of each equipment.

e) Membranes: Exergy destruction in the membranes is due to both stream exergy destruction (like part d) and composition change (like part c).

$$\dot{E}_{D,M} = \Delta Ex_{oc}(feed)-[\Delta Ex_{oc}(retentate)+\Delta Ex_{oc}(permeat)]+\dot{E}_{in}-\sum \dot{E}_{out} \qquad (16)$$

2.3 Membrane separation modeling

Polymer based membranes can be utilized in several gas separation operations such as hydrogen recovery, air separations and natural gas processing [17]. They have many prominent features like low energy and capital cost requirements, however, cannot be used to achieve high permeability and selectivity simultaneously. Moreover, although Inorganic membranes have high permeability and selectivity but they are so vulnerable during the operation and a defect such as a crack or gap between the particles would severely affect the membrane selectivity [18]. To take advantages of both polymeric and inorganic membranes, mixed matrix membranes (MMMs) are in favor. In fact, inorganic materials distributed in the organic polymer matrix enhance the separation performance of the membranes well beyond the intrinsic properties of the polymer matrix [19-20]. In this study a MMM based on poly(vinylidene fluoride) (PVDF) is used for gas separation. PVDF is a chemical resistance and durable semi-crystalline polymer which has flexible chains with high capability of membrane forming. Although PVDF is recognized as gas barrier material, in some points incorporation of porous fillers in polymer can improve its gas transport properties and turns it into a tool for gas separation. According to our previous work [20], PVDF comprising 10 wt% of CuBTC is a suitable MMM for Helium separation. The characteristics of this membrane are given in Table 2.

Table 2. Characteristics of the membrane [20]

PVDF-CuBTC-10%				
Permeability (Barrier)			Selectivity	
He	CH_4	N_2	He/ CH_4	He/ N_2
6.004	2.002	0.048	125.08	101.76

The flow diagram of separation process which is assumed to be cross flow and complete mixing is shown in Figure 3 [21].

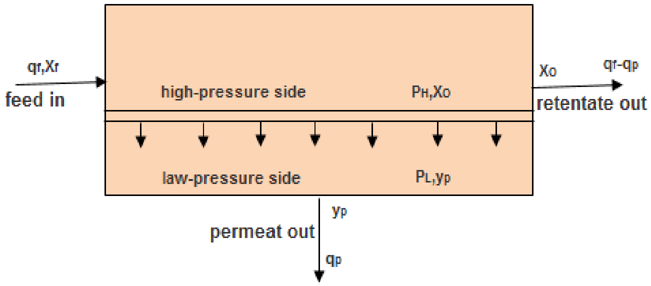

Figure 3 Process flow for separation of the membrane

In this study, y_{pHe}, which is the desired content of Helium in the permeate of the membrane is definite but y_{pCH_4}, y_{pN_2}, membrane area, flow rate of permeate and retentate and the molar composition of each one are unknown.The membrane area could be obtained from:

$$A_M = \frac{q_p y_{pHe} t}{P'_{He}[\frac{P_h}{1-\theta}(X_{fHe}-\theta y_{pHe})-P_l y_{pHe}]} \quad (17)$$

$$\theta = \frac{q_p}{q_f} \quad (18)$$

Molar fraction of any component like B in the permeate stream (y_{pB}) and in the retentate stream (X_{oB})can be determined by equations (19) and (20) respectively:

$$y_{pB} = \frac{P_h X_{fB} / 1\text{-}\theta}{q_p t / (p'_B A_M) + \theta P_h / 1\text{-}\theta + P_l} \quad (19) \qquad X_{oB} = \frac{1}{1-\theta} X_{fB} - \frac{\theta}{1-\theta} y_{pB} \quad (20)$$

Then an iteration procedure is used to find the membrane output parameters for the multi component mixture 36 separation. The flowchart of separation modeling flowchart is given in Figure 4.

Based on the above procedure and system configuration shown in Figure 2, the two stage membrane separation is modeled in MATLAB. It is assumed that permeate of the second membrane enriches the stream to 99% He.

2.4 Economic and environmental analysis

In order to evaluate economic aspects, capital and maintenance costs, operational costs and carbon dioxide production costs are provided and then all of the mentioned cost units are converted to $/s and added together and so, total cost rate of the whole system is obtained.

Cost data is often expressed as a power law of capacity multiplied by material, pressure and temperature factors.

$$Z_k = C_B(\frac{Q}{Q_B})^M f_M f_P f_T \quad (21)$$

where Z_k is the cost of equipment with capacity of Q, C_B stands for known base cost for equipment with capacity Q_B, M is a constant depending on equipment type and f_M, f_P and $f_T$are material, pressure and temperature capital cost factors respectively.

To calculate the capital and maintenance costs, estimated cost parameters for each component of the system is given in Table 3 and correction factors in Table 4 [22]. Having capital cost function for each component (Z_k), its capital cost rate (USD/unit of time) can be written as:

$$\dot{Z}_k = \frac{Z_k \times CRF \times \phi}{N \times 3600} \tag{22}$$

CRF is calculated by using the following formula [23]:

$$CRF = \frac{i(1+i)^n}{(1+i)^n - 1} \tag{23}$$

Table 3 Capital Cost-capacity function parameters [22]

Equipment	Capacity measure	Base size Q_B	Base cost C_B ($)	Cost exponent M
Pressure vessels	Mass (t)	6	$9.84x10^4$	0.82
Heat exchangers	Heat transfer area (m^2)	80	$3.28x10^4$	0.68
Compressors	Power (Kw)	250	$9.84x10^4$	0.46

Moreover, for expansion valve and membranes, cost functions are expressed by Equations (24) and (25) respectively.

$$Z_{EX} = 114.5 \times \dot{m}_r \quad (24) \qquad Z_M = A_M \times K_M \quad (25)$$

$\dot{m}_r$ is the fluid flow rate (kg/s) passing from the expansion valve and K_m is the membrane unit cost which is equal to 50 $/$m^2$.

The operational cost of the system is related to the electricity and natural gas (feed) consumption [24].

More electricity generation lead to more CO_2 emissions. Thus, in this paper CO_2 emission is considered as an important factor in modeling of the system. Amount of CO_2 emission generation by electricity consumption is as follows:

$$m_{co_2} = \mu_{co_2} \times S_{elec} \tag{26}$$

Table 4 Correction factors for estimating capital cost of the components [22]

Type	Range	Correction factor	Type	Range	Correction factor
Temperature (°C)	0-100	1.0	Pressure (bar)	50	1.5
Temperature (°C)	300	1.6	Pressure (bar)	100	1.9
Temperature (°C)	500	2.1	Material	Carbon steel	1.0
Pressure (bar)	0.1	1.3	Material	Aluminum	1.3
Pressure (bar)	0.5-7	1.0	Material	Stainless steel	2.4

Emission conversion factor value is equal to 0.968 kg kWh^{-1} [25]. CO_2 emission penalty cost is assumed as 90 US dollars per ton of CO_2 emissions [26].Therefore, the rate of CO_2 emission penalty cost is expressed by:

$$\dot{C}_{env} = \frac{\left(\frac{m_{co_2}}{1000}\right) \times c_{co_2}}{N \times 3600} \tag{27}$$

2.5 Optimization

2.5.1 Objective functions and design parameter

The two objective functions in multi-objective optimization considered for this study are exergy efficiency of the whole system (to be maximized) and the total cost rate of the whole system (to be minimized). Exergy efficiency as Objective function I is as follow:

$$OF_I: \Psi_{tot} = 1 - \frac{\dot{E}_{D,tot}}{\dot{E}_{in}}$$

where $\dot{E}_{D,tot}$ is the summation of exergy destruction rates of the system components and $\dot{E}_{in}$ is input exergy of the entire system.

Total cost rate as Objective function II is obtained from:

$$OF_{II}: \dot{Z}_k = \sum_k \dot{Z}_k + \dot{Z}_{op} + \dot{C}_{env}$$

The design parameter of the combined cryogenic-membrane system is the pressure of the feed which affects the exergy and cost of cryogenic system components as like as the ones for membrane system due to its effect on the pressure of stream 36.

2.5.2 Non-dominated sorting genetic algorithm

Srinivas and Deb [27] used NSGA sorting procedure as a ranking selection method, and a fitness sharing niching method to maintain stable sub-populations across the pareto front. Deb *et al.* [28] later developed NSGA to address three criticism of the approach: the lack of elitism, time complexity and needed for a sharing parameter for the fitness sharing niching method. Deb et al. introduced NSGA-II that contains a second-order sorting criterion named crowding distance which is faster and more flexible than NSGA. Figure 5 depicts the algorithm evolution procedure from the first step to the end.

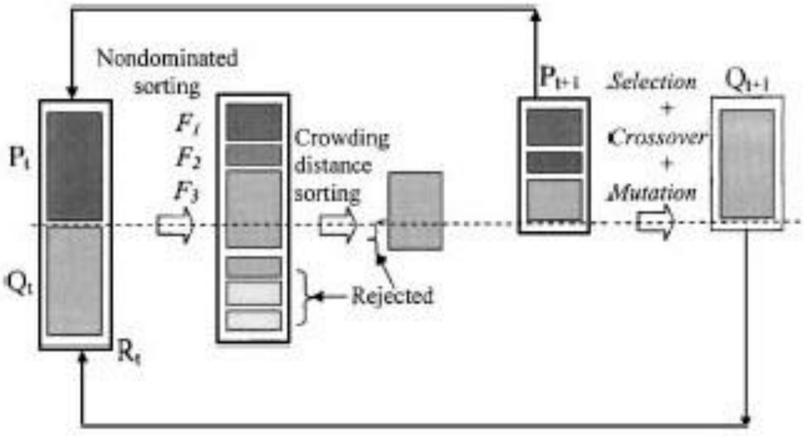

Figure 5 NSGA-II evolution procedure

The NSGA-II parameters for the model of this research are reported in Table 5.

Table 5. NSGA-II parameters for the model

Parameter	Value	Parameter	Value
Population size	80	Mutation percentage	0.4
Crossover percentage	0.8	Mutation range	0.02

3. Results and discussion

3.1 Model verification

All of the primary simulation by HYSYS and EES coding of cryogenic section are validated in comparison with the given data reported in reference 14. Moreover, the data reported in Table 2, are obtained from a membrane laboratory located in University of Tehran, Iran, which is also reported in reference 20.

Thermodynamics analysis issimple and precise with EES software (Engineering Equation Solver) because thermo-physical properties of fluids are available in the database of EES.

The feed gas of the studied plant contains 10 component, which all of their thermodynamic properties (i.e., enthalpy, entropy, specific heat, etc. in different temperatures and pressures) are accessible by EES. On the other hand, MATLAB has a powerful tool in Multi-objective optimization so that most of investigations in the field of the optimization of energy system are performed with MATLAB [29].

In this work, in order to take advantage of both EES and MATLAB, the analyzed and modeled system in EES is remodeled for MATLAB by regression in Excel. For this purpose, several runs are got from EES and the obtained data (more than 300 data series) are used for remodeling by regression in Excel. By doing this, a relation between the design parameter (pressure of the feed) and the two objective functions (exergy efficiency and total cost rate) is achieved to be used in NSGA-II algorithm. To evaluate the regression accuracy absolute fraction of variation(R^2), which is a common approach for this purpose is considered. R^2 could be calculated from the following equation [30]:

$$R^2=1-\left(\frac{\sum_i (T_{ri}-Q_i)^2}{\sum_i (Q_i)^2}\right) \qquad (28)$$

For this study, regression performance for feed pressure-exergy efficiency and feed pressure-total cost ratewas valid enough with R^2 values of 0.996 and 0.998 respectively.

3.2 Results of primary simulation

In the primary simulation of the system, the most important streams which are obtained after separations by flash tanks and membranes are shown in Table 6. In fact, the primary target of this project was to recover a gas stream containing 99% Helium (42) and LNG and fuel streams (12 and 35) from the feed gas.

Table 6 Characteristics of important streams of the combined cryogenic-membrane process

Fluid number	3	7	10	12	13	35	36	38	42	43
Phase	Vapor	Vapor	Vapor	Liquid	Vapor	Vapor	Vapor	Vapor	Vapor	Vapor
Temp.(°C)	-131.7	-133.3	-147.3	-159.9	-159.9	25	25	25	25	25
Pres.(kPa)	1080	760	310	120	120	2000	600	40	40	600
Flow rate (kg/s)	2.685	0.0485	0.327	2.184	0.217	0.592	0.00116	0.00112	0.0005	0.00062
Helium mol%	0.04	2.06	0.07	-	-	0.04	59.87	75	99	56.13
n-Hexane mol%	0.01	-	-	0.01	-	-	-	-	-	-
n-Pentane mol%	0.01	-	-	0.01	-	-	-	-	-	-
i-Pentane mol%	0.02	-	-	0.02	-	-	-	-	-	-
n-Butane mol%	0.18	-	-	0.23	-	-	-	-	-	-
i-Butane mol%	0.11	-	-	0.14	-	-	-	-	-	-
Propane mol%	0.53	-	-	0.67	-	-	-	-	-	-
Ethane mol%	1.6	0.02	0.01	2.02	-	0.01	-	-	-	-
Methane mol%	95.41	82.91	89.49	96.65	94.18	90.95	7.01	4	0.13	7.04
Nitrogen mol%	2.09	15.01	10.43	0.25	5.81	9	33.13	21	0.87	36.83

3.3 Optimization results

In this study, the decision variable (i.e., feed pressure) should be found in such a way to maximize the exergy efficiency (OF_I) and minimize the total cost rate (OF_{II}). It is assumed that the importance of each objective function is equal to the other one. The range of variation for feed pressure is varied from 1120 to 2625 kPa. NSGA-II parameters in the optimization process are given in Table 5. The results indicate that by increasing the exergy efficiency, the total cost rate increases. Table 7 reports the results of exergy efficiency and corresponding total cost rates after the optimization process.

Table 7 Results of optimization with NSGA-II

Parameter	Value	Parameter	Value
Maximum exergy efficiency (%)	43.92	Minimum exergy efficiency (%)	24.32
Total cost rate at maximum exergy efficiency points(USDs-1)	0.0825	Total cost rates at minimum exergy efficiency points(USDs-1)	0.0209

The user may select each of these points as the optimal point based on the desired purpose of optimization, however, usually the optimal point is the point which satisfies all objective functions and it is often placed between the first and the last points of the pareto frontier. In order to find the optimal point of optimization, Decision making is applied based on non-dimensionalizing of all objective functions. This is due to the fact that in most of the cases the units of objective functions are not the same. In this paper, TOPSIS method which is a technique for order preference by similarity to an Ideal solution is used. In this method, the shortest and farthest distances from the ideal point, are considered to choose the optimal solution [31].The following factor is defined for sorting the solutions:

$$Cl_i = \frac{d_i^-}{d_i^+ + d_i^-} \tag{29}$$

where d_i^- and d_i^+ are interval of optimization points from negative-ideal point and ideal point respectively.

Figure 6illustrates the pareto frontiers and TOPSIS selected optimal point.

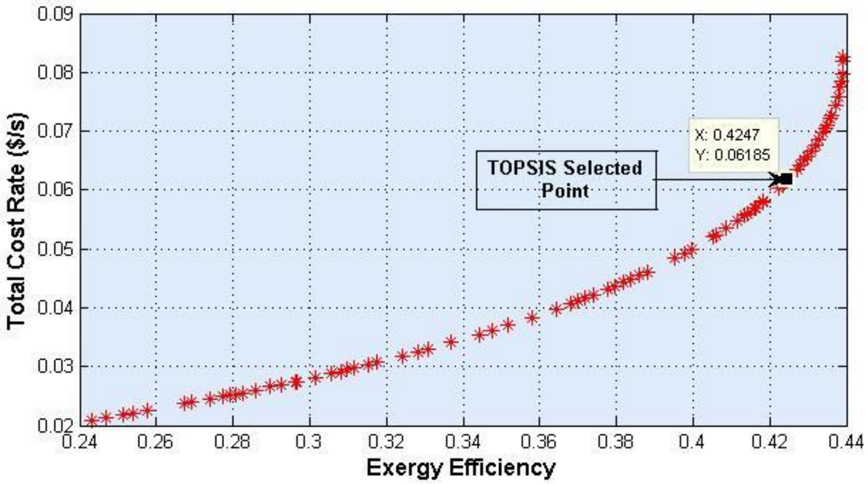

Figure 6 Pareto frontiers and TOPSIS selected optimal point

As it could be seen at the optimal point, exergy efficiency and total cost rate are 42.47% and 0.06185 $/s respectively. The corresponding optimal design parameter (feed pressure) is 2006 kPa and pressure of stream 36 is 1486 kPa. Table 8shows the components with the highest exergy destruction.

Table 8 Components with the highest exergy destruction at the optimal point

Equipment	Exergy destruction (kW)	Equipment	Exergy destruction (kW)
LNG liquefier	432	Second exchanger (Exchanger2)	286.9
Second expansion valve (VLV2)	415.3	Second fuel cooler (CL2)	191.9
Third expansion valve (VLV3)	258.5	Third fuel compressor (C3)	193.2

The results indicate that the highest exergy destruction occurs in the LNG liquefier. VLV2, Exchanger2, VLV3, C3 and CL2 are in the next orders. It should be also mentioned that, because very law flow rate of the feed stream enters to the membrane process section (about 0.04 of the feed), the membranes and other equipment of this section does not have a significant impact on total exergy efficiency. However, due to relatively high price of the membrane and high dependence of membrane area to the pressure of stream 36, the effect of membrane process section on the total cost rate of the combined system is noticeable. Figure 7 depicts variation of total membrane area by the pressure input to the membrane process section. As it is shown by increasing the inlet pressure, required membrane area decreases and so the fixed cost of membranes decreases, however, considering the combined cryogenic-membrane process as an integrated system and with respect to both exergy efficiency and total cost rate, the resulted inlet pressure to membrane process is 1486 kPa.

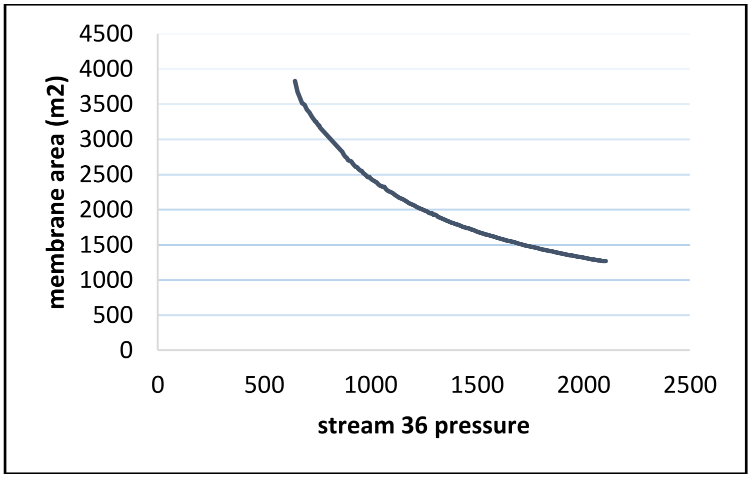

Figure 7 Variation of total membrane area by stream 36 pressure

4. Conclusion

A combined cryogenic flashing based system combined with a two stage membrane system for recovering Helium from natural gas was introduced. The proposed process can separate Helium gas, fuel gas and LNG streams from the feed. As Helium recovery processes are expensive and energy intensive, multi-objective optimization was carried out. The results indicated that when considering exergy efficiency and total cost rate of the combined system as objective functions with equal importance, 42.47% and 0.06185 \$/s were obtained at the optimal point. The feed gas pressure at this point was 2006 kPa and the inlet feed pressure to the membrane was 1486 kPa. Exergy analysis could help the designers for choosing the most exergy destructive components for improving the system performance. LNG liquefier, second and third expansion valves exergy destructions were the highest ones among the others. In addition it was found that although membrane section did not have an important role on total exergy efficiency due to low flow rate of inlet stream, but relatively high price of membrane affected the total cost rateof the system. At the optimal point, the required membrane area was 1710 m^2.

Evaluating other type of membranes and other configurations for membrane process and for the whole system may be useful for further investigations.

Nomenclatures

$\dot{E}$	*Exergy flow rate (KW)*	*CRF*	*Capital recovery fact*
i	*Annual intrest rate*	*h*	*Specific enthalpy (kJ/k)*
N	*Annual operational hours (hr)*	*He*	*Helium*
$\dot{Q}$	*Heat transfer rate (KW)*	$\dot{m}$	*Mass rate (kg/S)*
s	*Specific entropy (kJ/kg)*	*P*	*Pressure (kPa)*
η	*Efficiency*	*Ph*	*Physical*
Φ	*Maintenance factor*	*T*	*Temperature (C)*
FT	*Flash Tank*	*env*	*Environment*
C	*Compressor*	ψ	*Exergy efficiency*
CL	*Cooler*	*VLV*	*Valve*
liq	*Liquefier*	*EES*	*Engineering equation software*
e	*Exit*	*f*	*Factor*
R	*Gas Constant*	*i*	*Inlet*
n	*Number of moles*	*Op*	*Operational*
μ	*emission conversion factor*	*O*	*Outlet*
m_{CO_2}	*Mass of dioxide carbon*	*M*	*Membrane*
c_{co_2}	*emission penalty cost per Ton*	*S*	*Annual consumption*
t	*Membrane thickness*	$\dot{C}_{env}$	*Rate of emission penalty cost*
Ph	*Higher pressure*	*q*	*Flow rate*
Z	*Cost ($)*	P_l	*Lower pressure*
$\dot{Z}$	*Investment cost rate ($/S)*		

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Corresponding author: E-mail: tavassolia@khayam.ut.ac.ir, Phone: (98)-21-61113643, Fax: (98)-21-66495291