

Research paper

Energy, exergy and economic analyses of two modified and optimized small-scale natural gas liquefaction (LNG) cycles using N₂ and N₂/CH₄ refrigerants with CO₂ precooling



Sepehr Sanaye*, Seyed Milad Shams Ghoreishi

Energy System Improvement Laboratory (ESIL), School of Mechanical Engineering, Iran University of Science & Technology (IUST), Iran

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ABSTRACT

Natural gas liquefaction cycles require high amounts of shaft power and any effort for decreasing power consumption of these cycles is a subject of interest. Among natural gas liquefaction cycles, N₂ and N₂/CH₄ cycles with CO₂ pre-cooling are attractive due to their simplicity. The novel work here is that two above cycles are first modified. Then by choosing the ratio of power consumption to the liquefied gas molar flow rate (Specific Energy Consumption, SEC) as the objective function and 13 design parameters for N₂-CO₂ cycle and 12 design parameters for N₂/CH₄-CO₂ cycle, these cycles are optimized. Results of the above procedure showed decrease in both SEC and the total annual cost in comparison with those for similar cycles published in open literature. Two modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ liquefaction cycles showed 5.25% and 3.62% decrease in specific energy consumption and 9.4% and 16% decrease in exergy destruction respectively. The total cost of two above cycles also decreased for 0.5% and 1.2% respectively.

1. Introduction

Natural gas (NG) is a relatively clean and important fossil fuel with many applications [1]. Natural gas is being used in industrial processes such as heating, power generation and in production of a wide range of chemicals. It is also being used in residential applications such as space heating, domestic hot water and cooking. Moreover, among three common fuels used for electric power generation (coal, oil and NG), NG is the cleanest fuel due to its lowest emission of CO₂ per unit of energy produced [2]. Natural gas at atmospheric pressure can be liquefied when its temperature is cooled below −161 °C [3]. Liquefied natural gas (LNG) is a form of natural gas which contains more than 98% methane [4].

LNG plants have intensive power consumption and they are increasing in number due to the growing demand for natural gas [5]. LNG may be transported by either pipelines or liquefied natural gas carriers. Its pipeline transportation cost is low, however it needs huge initial investment. For small gas fields and long-distance transportation, natural gas could be liquefied and transported by LNG carriers to reduce the cost.

LNG is odorless, clear and nontoxic liquid with a density of 450 kg/m³. Liquefaction of gas reduces its volume to 1/600 of the initial gas

state, which makes the transportation economically more beneficial, while this reduction is 1/200 and 1/250 in the case of LPG and CNG products respectively [6].

There are various natural gas liquefaction processes including cascade processes [7], mixed (made up of combination of nitrogen, methane, ethane and propane as refrigerant [8]) refrigerant expansion processes such as N₂/CH₄ cycle as well as N₂ expansion processes named Nitrogen expansion cycle [9].

Among above types of liquefaction processes, N₂ expansion process is recommended for small scale LNG plants [10] due to its faster startup and more convenient maintenance. However this problem has a disadvantage which is having high SEC value [11]. N₂/CH₄ which is a mixed refrigerant expansion process, has also a simple arrangement and low maintenance cost.

As the natural gas liquefaction process has high power consumption, performance improvement in these cycles, reduces both power consumption and CO₂ emissions which are produced during electricity generation in Power Plant.

There are several methods used for improving these processes.

The first method which can be used for both N₂ and N₂/CH₄ cycles is changing operating parameters (modifying cycle pressure, temperature, molar flow rate values) to reduce SEC value.

* Corresponding author at: Energy Systems Improvement Laboratory, School of mechanical Engineering, Iran University of Science and Technology, Narmak, Tehran 16488, Iran.

E-mail addresses: sepehr@iust.ac.ir (S. Sanaye), Qoreishi.m@gmail.com (S.M. Shams Ghoreishi).

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Nomenclature

ΔE_x	exergy losses (kJ)
η	efficiency
At	heat transfer surface (m ²)
E	specific exergy
F	flow
H	enthalpy (kJ)
h	specific enthalpy (kJ/kmol)
I	irreversibility
i	interest rate
Is	isentropic
N	system life time (year)
P	pressure (kPa)
S	entropy (kJ/K)
s	specific entropy (kJ/kmol/K)
T	temperature (K)
T	temperature (°C)
Q	heat flow (kW)
q	molar flow (kmol/h)
W	power (kW)
S	separator

Abbreviations

C3	propane
CNG	compressed natural gas
comp	compressor
Cv	control volume
COP	ratio of useful cooling provided to work required
CRF	capital recovery factor

E	compressor or expander or water cooler
exp	expander
GA	genetic algorithm
HEX	heat exchanger
HP	horse power
LNG	liquefied natural gas
LPG	liquid petroleum gas
mech	mechanical
MIX	component for combining two streams
MR	mixed refrigerant cycle
MR-C3	mixed refrigerant cycle with propane pre-cooling
MTDA	minimum temperature difference approach
N ₂ /CH ₄ -CO ₂	mixed refrigerant of nitrogen and methane expansion cycle liquefaction process with carbon dioxide pre-cooling
N ₂ -CO ₂	nitrogen expansion cycle liquefaction process with carbon dioxide pre-cooling
N ₂ -R410a	N ₂ -R410a nitrogen expansion cycle liquefaction process with R410a pre-cooling
NA	not available
NCE	N ₂ /CH ₄ expansion cycle
NE	nitrogen expansion cycle
NG	natural gas
PEC	purchase equipment cost
SEC	specific energy consumption
SP	skid-mounted Package
TEE	component for separating one stream into two streams
VLV	throttling valve
UEC	unit energy consumption
WC	water cooler

Li and Ju [10] designed a N₂ cycle for an offshore natural liquefaction plant and managed to improve the liquefaction process by modifying its operating parameters according to the offshore associated gas resources in South China Sea and compared the results with mixed refrigerant cycle with and without propane precooling. Results showed that in the modified N₂ cycle, SEC value (0.4 kWh/Nm³) was higher than two MR-C3 and MR cycles (0.238 and 0.263 kWh/Nm³ respectively). However, N₂ cycle is a relatively appropriate cycle for LNG plants due to its compactness, easy operation, better performance under different feed gas composition and less area required for decking. Cao et al. [12] optimized N₂/CH₄ and other mixed refrigerant expansion process without pre-cooling cycle. Results showed that SEC value of N₂/CH₄ (93.2 kW/mol/s) was lower than that for mixed refrigerant expansion processes without pre-cooling (122.3 kW/mol/s). Then he compared three cycles of N₂/CH₄ cycle, mixed refrigerant cycle with propane pre-cooling and mixed refrigerant cycle in skid-mounted Package [13]. As the result, they concluded that the mixed refrigerant cycle in skid-mounted package was the most suitable due to its acceptable SEC value and low equipment number. Tianbiao et al. [14] also used a comprehensive optimization method for parallel N₂ and modified MR cycles and compared their SEC values. Results showed that SEC value for the modified MR process (0.411 kWh/kg) was lower than that for parallel N₂ cycle (0.618 kWh/kg). Qayyum [15] et al. used highly efficient hybrid modified coordinate descent (HMCD) algorithm for improving operating parameters of MR cycle. Result showed the enhanced COP value for about 34.7% as compared to the base case.

Modifications in equipment arrangement for N₂ and N₂/CH₄ cycles are also reported as the second method for reducing the SEC value by decreasing temperature difference in heat exchangers. For example, Li and Ju [10] and Gao et al. [16] proposed cascade configuration for N₂ cycle with 50 °C temperature differences in heat exchangers (which reduced the efficiency of LNG production cycle).

Adding pre-cooling to the main refrigeration cycle is the third method of reducing SEC value in a N₂ cycle, however pre-cooling increases the cycle complexity and investment cost. Li et al. [17] proposed N₂ cycle with R410a as refrigerant for pre-cooling cycle (N₂-R410a) in an offshore natural gas liquefaction plant. Results showed that the thermodynamic performance of the N₂-R410a was better than that of N₂ cycle without pre-cooling. He and Ju [11] also presented N₂ cycle with two separated pre-cooling cycles with propane (N₂-C₃) and R410a (N₂-R410a) refrigerants, and compared the SEC value for N₂ cycle with and without pre-cooling. Results revealed that the lowest SEC value occurred for N₂-R410a (with SEC value of 0.3607 kWh/Nm³) and the next best option was N₂-C₃ cycle (with SEC value of 0.3734 kWh/Nm³).

CO₂ pre-cooling due to safety, environmentally friendly, being non-toxic and non-combusted was important. Furthermore CO₂ pre cooling cycle had simple reduction and storage advantages.

Zongming et al. [18] compared N₂-CO₂ (N₂ with CO₂ Precooling) and N₂/CH₄ mixed refrigerant cycle without pre-cooling cycle. The comparison showed N₂-CO₂ with SEC value of 9.90 kW/kmol/h had lower SEC value compared to N₂/CH₄ cycle with SEC value of 17.68 kW/kmol/h, however it had two additional compressors which increased the complexity (12 versus for 10 for number of main equipment) for N₂-CO₂ and N₂-CH₄ respectively.

Adding pre-cooling has been also used in mixed refrigerant cycles such as N₂/CH₄ for reducing SEC value. Zongming et al. [18] also added a CO₂ precooling cycle to the main heat exchanger (HEX-101) of N₂/CH₄ mix refrigerant cycle which decreased SEC value. In their research they shifted a apart of cooling capacity from cooling cycle to the pre cooling cycle which increased the cycle efficiency.

Ding et al. [19] also proposed N₂/CH₄ with CO₂ and propane pre-cooling cycles. Results showed that SEC value reduced to 9.11 and 6.73 kWh/kmol for N₂/CH₄-CO₂ and N₂/CH₄-C₃ cycles respectively. As a

result, CO₂ pre-cooling cycle is considered as an economic cycle with low cost and acceptable cooling capacity.

From economic point of view, reducing the number of equipment, simplifying the cycle and optimization are all important in decreasing the investment and operating costs which results in rising the net profit. Tianbiao [20] optimized two cycles of modified mixed refrigerant liquefaction process and parallel nitrogen expansion liquefaction process by using either objective function of SEC value or total cost which both should be minimized. Results showed SEC value and total cost in the modified mixed refrigerant liquefaction were 0.411 kWh/kg and \$6,126,133 respectively for the modified cycle in comparison with 0.618 kWh/kg and \$8,379,177 respectively for the base cycle.

There are still challenges in modification of liquefaction processes with CO₂ pre-cooling cycle including efforts to decrease the temperature difference of hot and cold side fluids in heat exchangers, which causes reduction in exergy destruction in liquefaction operation as well as reduction in power consumption and efficiency.

In this paper two improved N₂-CO₂ and N₂/CH₄-CO₂ expansion liquefaction cycles with CO₂ pre-cooling are proposed. In the first step, both cycles were modified by new arrangement of equipment or adding equipment such as new heat exchangers or distributors as well as change in operating parameters such as molar flow rates in the main LNG refrigeration cycles. In the second step, by choosing specific energy consumption as an objective function and 13 cycle design parameters for the modified N₂-CO₂ cycle and 12 design parameters for the modified N₂/CH₄-CO₂ cycle, both cycles were modeled and optimized. Results showed that the above modified and optimized cycles had lower specific energy consumption (SEC) at similar liquefaction rate in comparison with those reported in Refs. [18] and [19].

Furthermore exergy destruction decreased in main heat exchangers of both above cycles. This reduction in exergy destruction was mainly due to closer hot and cold temperature curves in main heat exchangers which could be gained from optimization results.

Finally, by economic analysis of the above cycles, it was observed that annual investment and maintenance costs increased due to using bigger heat transfer surface area with lower hot and cold temperature differences. However, it should be noted that the total annual cost decreased due to high reduction in operating cost in comparison with that for cycles discussed in Refs. [18,19].

2. System description

N₂ expansion liquefaction cycle with CO₂ pre-cooling (N₂-CO₂) is shown in Fig. 1a. This figure shows the cycle investigated in Ref. [11] which a heat exchanger (HEX-101) and a distributor (TEE-100) are added into the cycle and its operating conditions are changed. In this cycle the feed gas after pretreatment of natural gas, passes through heat exchanger HEX-102 where its temperature decreases to about −30 °C. In HEX-102 a part of returning N₂ from HEX-103 and the separated heavy hydrocarbons in S-100 were cold streams. After passing through liquid-vapor separator S-100, the vapor part cools down further in HEX-103 to reach −131.7 °C. Then the feed gas pressure and temperature reduce after expansion valve VLV-102 to LNG storage pressure of 200 kPa and temperature of −150.9 °C. At this step, LNG product is separated out in vapor-liquid separator S-101 and the flash gas is sent to HEX-101 for N₂ pre-cooling. In this heat exchanger N₂ is cooled by four streams including CO₂ refrigerant (stream 307), flash gas (stream 111), a part of returning N₂ (stream 211) and separated heavy hydrocarbon (stream 106).

For N₂ cycle, nitrogen undergoes two stages of compression in C-201 and C-202 compressors and two cooling processes in WC-100 and WC-101 by water-coolers. N₂ cools down further in HEX-101 by carbon dioxide pre-cooling process after which its temperature drops to −51.9 °C in a flashing process. After undergoing a single stage of expansion in E-201, N₂ temperature decreases to −134.1 °C which would be used as a cooling fluid in HEX-101, HEX-102 and HEX-103. In this cycle, a TEE was used as a divider for making two stream of refrigerant which each part passed through HEX-101 and HEX-102. This change in refrigerant mass flow rate in heat exchangers to decrease the temperature difference in hot and cold composite curves in HEX-101 are shown in Figs. 2a and 2b.

In pre-cooling cycle, pressure of CO₂ increases by two stages of compression through passing in C-301 and C-302 compressors and its temperature drops to about 35 °C by passing through WC-102 and WC-103 water-coolers. Finally, the CO₂ pressure decreases in throttling valve VLV-301 to be used as cold fluid in HEX-101 and HEX-102.

Fig. 1b also illustrates the modified N₂/CH₄ expansion liquefaction cycle with CO₂ pre-cooling (N₂/CH₄-CO₂). In this cycle, the feed gas, after pretreatment, is cooled to −147.6 °C in HEX-102. Then it passes through the throttling valve VLV-101 to reduce its pressure and temperature to the pressure of LNG storage tank of 200 kPa at −152.9 °C respectively. Finally, LNG product separates in vapor-liquid separator S-

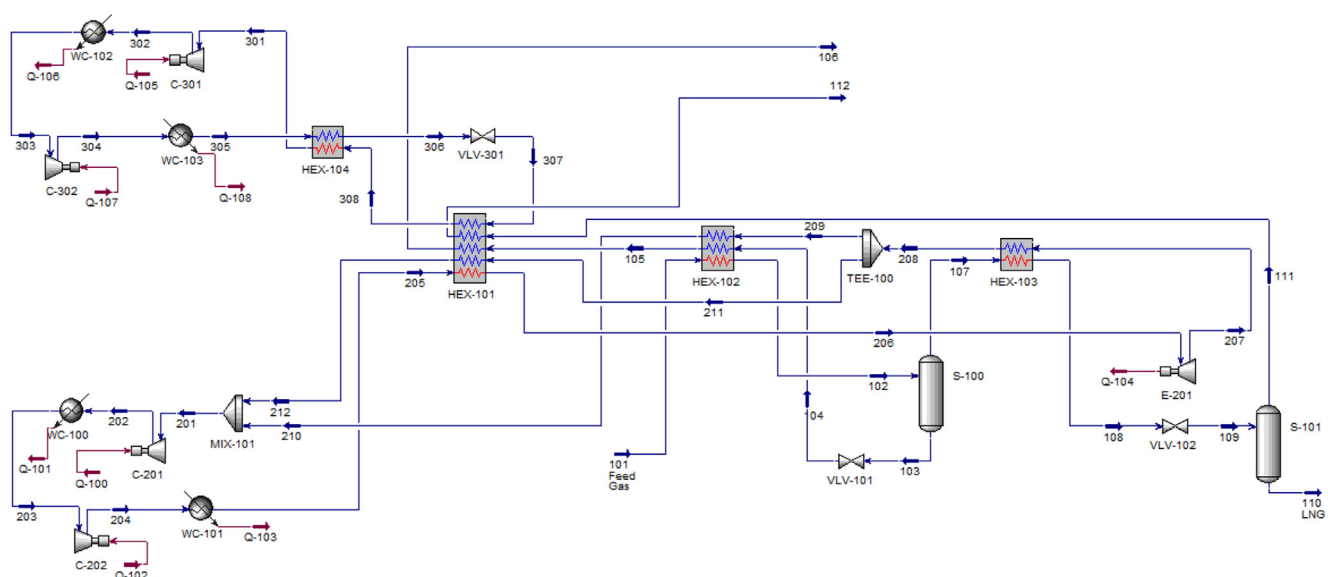


Fig. 1a. The modified N₂ cycle with CO₂ pre-cooling liquefaction cycle (N₂-CO₂).

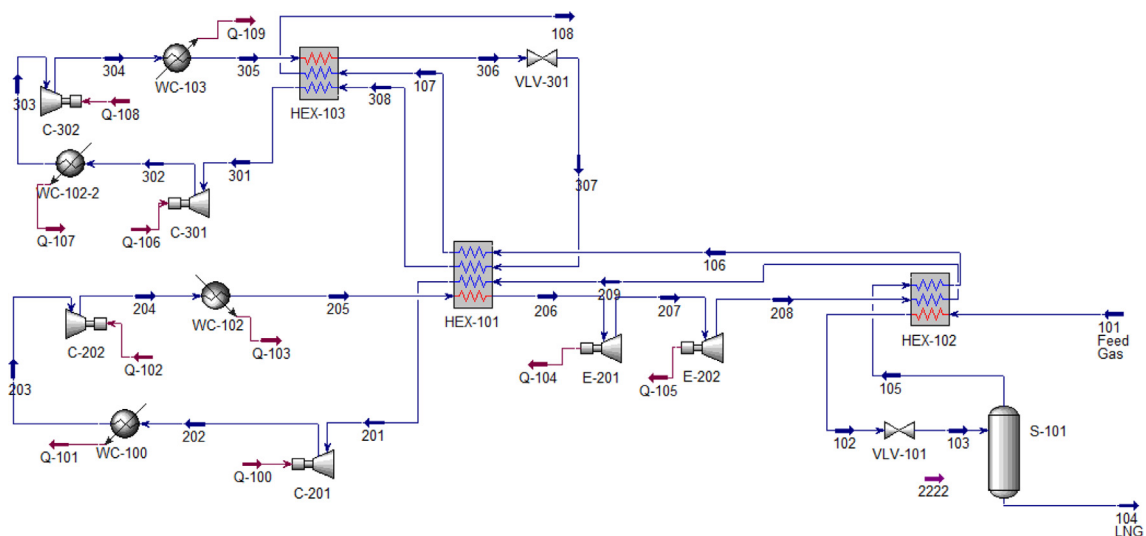


Fig. 1b. The modified mixed refrigerant (nitrogen and methane) cycle with CO₂ pre-cooling liquefaction cycle (N₂/CH₄-CO₂).

101. The vapor exiting from the separator then enters HEX-102 and HEX-101 as cold fluid for cooling feed gas and mixed refrigerant streams.

In N₂/CH₄ refrigeration cycle, the mixed refrigerant undergoes two stages of compression in C-201 and C-202 compressors and two cooling processes in WC-100 and WC-101 water coolers. The refrigerant then further cools down in HEX-101 to -30.0°C . Afterward, it undergoes two single stage expansion in E-201 and E-202 to reach a low pressure/temperature condition of 233 kPa/ -149.6°C . This final state of refrigerant could be used as cold fluid in HEX-102.

The pre-cooling system of this cycle is as the same as the one for N₂ with CO₂ pre-cooling, except that one of heat exchanger is omitted.

HEX-101 in both N₂-CO₂ and N₂/CH₄-CO₂ cycles was as a key component through which refrigerants, N₂ or N₂/CH₄ and CO₂ as well as natural gas reflux were flowing through (Figs. 1a, 1b). Hot and cold composite curves for the main heat exchanger HEX-101 in two above cycles are shown in Figs. 2a and 2b. In HEX-101, the high pressure refrigerant is hot fluid while CO₂ and natural gas reflux are cold fluids. To limit the required heat transfer surface area, a minimum approach

temperature (ΔT_{\min}), of 2°C was selected in heat exchangers.

Due to this close temperature variation of hot and cold fluids as is illustrated in Figs. 2a and 2b, the exergy destruction in the main heat exchanger HEX-101 was low. However, for closer approach temperatures, heat transfer surface area and its corresponding cost increase.

3. Exergy analysis

Exergy is defined as the maximum useful work during a process that brings the system into equilibrium with surroundings. To improve (reduce) irreversibility in processes, exergy analysis can be applied. This can be performed by defining exergy efficiency which shows the total system exergy destruction in a process and actually computes the effectiveness of a system relative to its performance in reversible conditions [21].

For a control volume, the exergy conservation equation is [22]:

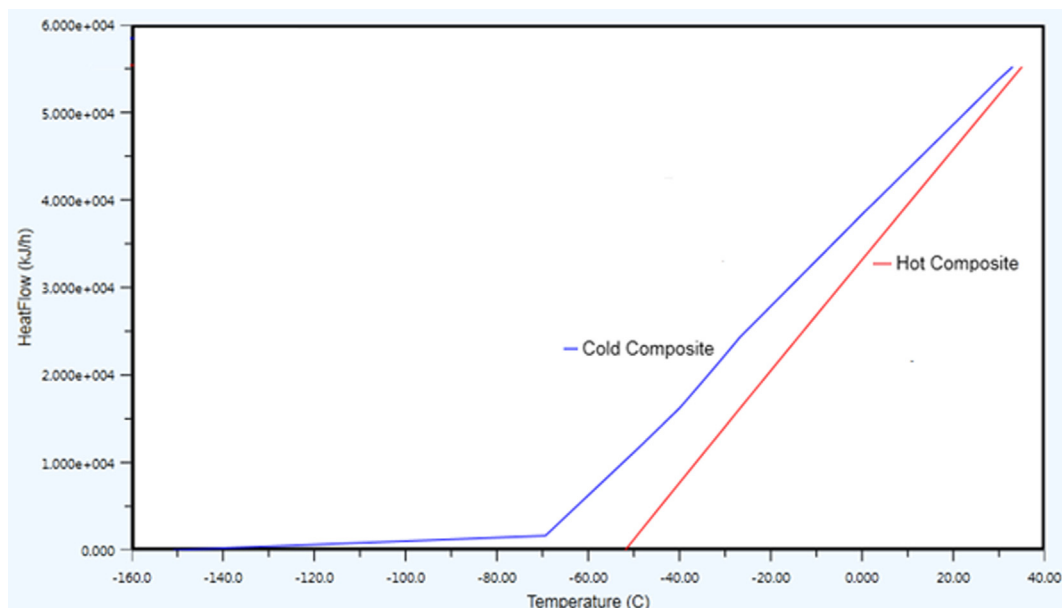


Fig. 2a. Hot and cold composite curves for heat exchanger HEX-101 in N₂-CO₂ cycle.

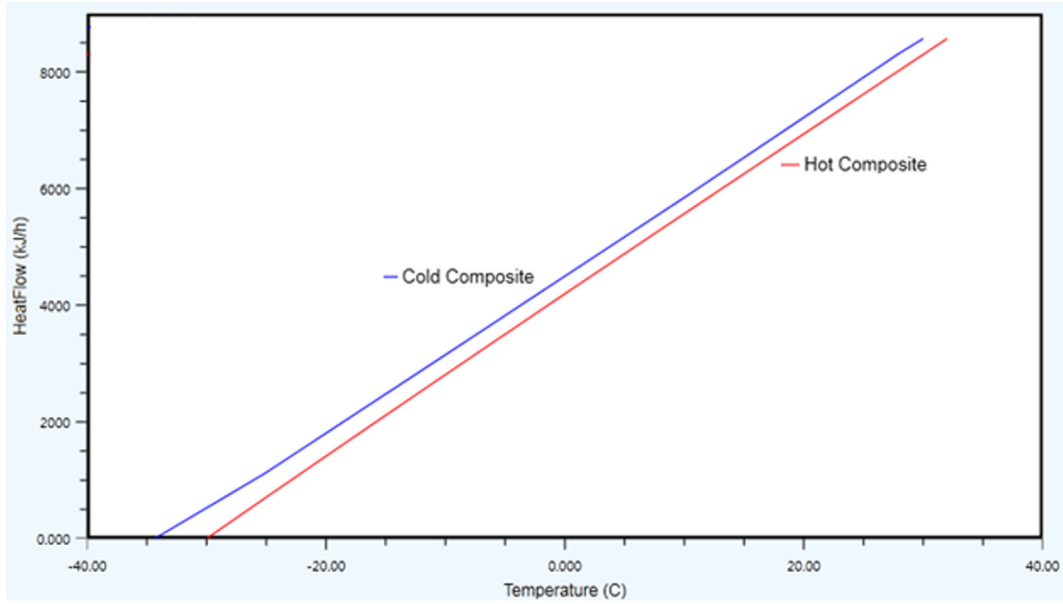


Fig. 2b. Hot and cold composite curves for heat exchanger HEX-101 in N₂/CH₄-CO₂ cycle.

$$\frac{dE_{cv}}{dt} = \sum_j \left(1 - \frac{T_0}{T_j}\right) \dot{Q}_j - \left(\dot{W}_{cv} - p_0 \frac{dV_{cv}}{dt}\right) + \sum_i \dot{m}_i e_{fi} - \sum_i \dot{m}_e e_{fe} - \dot{I}_{cv} \quad (1)$$

where

$$e_f = (h - h_0) - T_0(s - s_0) + \frac{V^2}{2} + gz \quad (2)$$

$\left(1 - \frac{T_0}{T_j}\right) \dot{Q}_j$ is exergy transferred by heat transfer. \dot{Q}_j is the heat rate transferred from the outer surfaces with temperature T_j , \dot{W}_{cv} is the work transfer rate transferred from boundaries of control volume and $\left(\dot{W}_{cv} - p_0 \frac{dV_{cv}}{dt}\right)$ is exergy transfer rate by both work transfer rate \dot{W}_{cv} and flow work $p_0 \frac{dV_{cv}}{dt}$ where $\frac{dV_{cv}}{dt}$ is time rate of change in volume.

Thus for an equipment with steady state steady flow conditions with negligible kinetic and potential exergy, Eq. (1) reduces to:

$$0 = \sum_j \left(1 - \frac{T_0}{T_j}\right) \dot{Q}_j - (\dot{W}_{cv}) + \sum_i \dot{m}_i e_{fi} - \sum_i \dot{m}_e e_{fe} - \dot{I}_{cv} \quad (3)$$

where exergy destruction per unit mass flow rate ($\sum_i \dot{m}_i = \sum_i \dot{m}_e = \dot{m}$) is:

$$\frac{\dot{I}_{cv}}{\dot{m}} = \sum_j \left(1 - \frac{T_0}{T_j}\right) \dot{Q}_j - \left(\frac{\dot{W}_{cv}}{\dot{m}}\right) + \frac{\sum_i \dot{m}_i e_{fi} - \sum_i \dot{m}_e e_{fe}}{\dot{m}} \quad (4)$$

Thus in compressor and turbine, without heat transfer from outer boundaries, exergy destruction for one inlet and one outlet can be explained as:

$$\frac{\dot{I}_{cv}}{\dot{m}} = -\left(\frac{\dot{W}_{cv}}{\dot{m}}\right) + (e_{f1} - e_{f2}) \quad (5)$$

In water coolers and heat transfer equipment, without heat transfer in boundaries, exergy destruction for one inlet and one outlet can be explained as:

$$\frac{\dot{I}_{cv}}{\dot{m}} = \sum_j \left(1 - \frac{T_0}{T_j}\right) \dot{Q}_j + (e_{f1} - e_{f2}) \quad (6)$$

T_j denotes the temperature on the boundary where heat transfer occurs and T_0 is the ambient temperature.

For heat exchangers, without work and heat transfer from outer boundaries, exergy destruction per unit mass flow rate is estimated

from:

$$\frac{\dot{I}_{cv}}{\dot{m}} = \frac{\sum_i \dot{m}_i e_{fi} - \sum_i \dot{m}_e e_{fe}}{\dot{m}} \quad (7)$$

For exergy destruction in valves also Eq. (7) may be used with one inlet and one outlet.

4. Economic analysis

The economic analysis of N₂-CO₂ and N₂/CH₄-CO₂ cycles was also performed to estimate the total investment and operational costs. As these costs relate to specifications of equipment and their operating conditions, thus total annual cost can be considered as a criterion for choosing the optimum cycle in both technical and economical points of views.

4.1. Investment cost

Investment costs for equipment are estimated based on cost functions presented below.

For compressors [23]:

$$PEC_{comp} = \frac{C_1 \times \dot{m}}{C_2 - \eta_{is}} \times \ln\left(\frac{P_{out}}{P_{in}}\right) \times \frac{P_{out}}{P_{in}} \quad (8)$$

where \dot{m} is the mass flow rate, η_{is} is the isentropic efficiency and C_1 and C_2 are constants equal to 573 \$/kg/s and 0.8996, respectively.

$$\eta_{is} = 0.85 - 0.046667 \times \frac{P_{out}}{P_{in}} \quad (9)$$

For heat exchanger [24]:

$$PEC_{heat-exchanger} = A_f \times \{C_a + C_b A_f^c\} \quad (10)$$

where A_f is the heat transfer surface which is estimated by assuming typical average value for overall heat transfer coefficient, U [25]. A_f , C_a , C_b and c are constants equal to 0.322, 30000, 750 and 0.8, respectively.

For expander [26]:

$$PEC_{expander} = 1000 \times 0.378 \times (HP)^{0.81} \quad (11)$$

where $HP = \frac{W_{exp}}{\eta_{mech} \times 735.5}$

η_{mech} is the mechanical efficiency of the expander.

4.2. Annual investment, maintenance and total costs

Operational and maintenance costs are annual. However, investment cost should be paid when one is purchasing equipment (purchasing equipment cost, PEC). To determine the annual investment cost during the system lifetime a coefficient (capital recovery factor, CRF) is defined as [27]:

$$CRF = \frac{i(1+i)^n}{(1+i)^n - 1} \quad (12)$$

where i is the interest rate and n is the system lifetime.

The product of capital recovery factor (CRF) and the cost of purchasing equipment (PEC), results annual capital recovery cost, CRC:

$$CRC = CRF \cdot PEC \quad (13)$$

For predicting annual maintenance and repairing cost, a coefficient of 1.06 was considered for PEC [28]. Thus total annual cost included the capital recovery cost (for investment and maintenance costs) and the annual operating cost.

5. Process optimization

5.1. Objective function, design variables and constraints

In this work, electrical power consumption per molar flow rate of LNG was selected as the objective function, thus:

$$f(x) = \min \left(\frac{W_{net}}{q_{LNG}} \right) \quad (14)$$

where W_{net} is the net power consumption.

$$W_{net} = \sum W_{compressor} - \sum W_{expander} \quad (15)$$

Design variables in N_2 -CO₂ and N_2 /CH₄-CO₂ are listed in Table 1.

In N_2 -CO₂ cycle 13 design parameters including, temperature of heavy hydrocarbons before passing through heat exchanger HEX-101 (t-105), minimum, intermediate and maximum pressure of compressors in refrigeration cycle (p-201, p-202, p-204), molar flow rate of refrigeration cycle (q-201), temperature of refrigerant after passing HEX-101 (t-206), distribution of molar flow rate in TEE-100 (q-209), minimum, intermediate and maximum pressure of pre-cooling cycle (p-301, p-302, p-304), molar flow rate of pre-cooling cycle (q-301), and temperature of pre-cooling cycle after passing HEX-101 and HEX-104 (t-306, t-308), are assumed as design variables.

In N_2 /CH₄-CO₂ cycle 12 design parameters including, minimum, intermediate and maximum pressure of compressors in refrigeration cycle (p-201, p-202, p-204), molar flow rate of refrigeration cycle (q-201), temperature of refrigerant after passing HEX-101 (t-206), intermediate pressure of expanders in refrigeration cycle (p-207), backflow

temperature of boiled off gas after passing HEX-102 (t-106), minimum, intermediate and maximum pressure of pre-cooling cycle (p-301, p-302, p-304), molar flow rate of pre-cooling cycle (q-301), and temperature of pre-cooling cycle after passing HEX-101 (t-306), are used as design variables.

Minimum temperature approach in heat exchangers was also selected as a constraint:

$$\min \text{ approach}(HEX - (i)) \geq 2(i = 101, 102, 103) \quad (16)$$

6. Results and discussion

6.1. Validation of modeling results

Modeling and optimizing of the N_2 -CO₂ and N_2 /CH₄-CO₂ cycles are conducted by Aspen HYSYS software [29]. This process simulation software package with the use of thermodynamic models for equipment, predicts the performance of processes in various input conditions. Peng Robinson equation [30] is adopted to compute thermodynamic properties such as enthalpy and entropy. This equation is a good standard database for pure and mixed hydrocarbons and their thermodynamic properties such as pressures and temperatures.

To validate the modeling results, the simulation output in this study were compared with those for cycles proposed in Refs. [18] and [19] for the same input values (Tables 2a and 2b).

Simulation results in this study for cycles proposed in Refs. [18] and [19] and those reported values in those references are also listed in Table 3 for N_2 -CO₂ and N_2 /CH₄-CO₂ cycles. The comparison of results showed maximum 4% difference which is acceptable in engineering computing.

6.2. Optimization results

After modifying N_2 -CO₂ and N_2 /CH₄-CO₂ cycles, optimization was performed by the use of HYSYS optimizer with “Mixed” method, with tolerance (5e-4) and maximum 200 iteration.

The ratio of power consumption to liquefied gas molar flow rate was chosen as an objective function and 13 cycle design parameters for modified N_2 -CO₂ cycle and 12 cycle design parameters for N_2 /CH₄-CO₂ cycle were selected for optimization procedure. At steady-state condition, HYSYS conducted optimization for N_2 -CO₂ and N_2 /CH₄-CO₂ cycles. The operating conditions at the optimum point (when the minimum value of objective function was reached) are listed in Tables 4a and 4b. Liquefaction rate, SEC and number of equipment at the optimum point are listed in Table 5. The optimized N_2 -CO₂ liquefaction cycle had specific energy consumption (SEC) value of 9.38 kWh/kmol at liquefaction rate of 0.77 which shows about 5.25% reduction in specific energy consumption (SEC) value at same liquefaction rate

Table 1
Design parameters (decision variables) in optimizing of N_2 -CO₂ and N_2 /CH₄-CO₂ processes.

N_2 -CO ₂ cycle		N_2 /CH ₄ -CO ₂ cycle	
Variable	Description	Variable	Description
t-105	Temperature of heavy hydrocarbons between HEX-101 and HEX-102	p-201, p-202, p-204	Minimum, intermediate and maximum pressure of compressors in refrigeration cycle
p-201, p-202, p-204	Minimum, intermediate and maximum pressure of compressors in refrigeration cycle	q-201	Molar flow rate of refrigeration cycle
q-201	Molar flow rate of refrigeration cycle	t-206	Temperature of refrigerant after passing HEX-101
t-206	Temperature of refrigerator after passing HEX-101	p-207	Intermediate pressure of expanders in refrigeration cycle
q-209	Distribution of molar flow rate in TEE-100	t-106	Backflow temperature of boiled off gas after passing HEX-102
p-301, p-302, p-304	Minimum, intermediate and maximum pressure of pre-cooling cycle	p-301, p-302, p-304	Minimum, intermediate and maximum pressure of pre-cooling cycle
q-301	Molar flow rate of pre-cooling cycle	q-301	Molar flow rate of pre-cooling cycle
t-306	Temperature of pre-cooling cycle after passing HEX-101	t-301	Temperature of pre-cooling cycle after passing HEX-101
t-308	Temperature of pre-cooling cycle after passing HEX-104		

Table 2a

Input temperatures, pressures and efficiencies for two existing cycles N_2 -CO₂ [18] and N_2 /CH₄-CO₂ [20].

Parameter	Value	
	N_2 -CO ₂ cycle [18]	N_2 /CH ₄ -CO ₂ cycle [19]
Feed Gas Pressure (kPa)	4800	5000
Feed Gas Temperature (C)	32	30
Feed Gas Flow Rate (kmol/h)	4	1
Temperature After Water Cooler (C)	35	32
Temperature for cold energy recovering backflow flashed gas	30 °C	30 °C
Ambient temperature	25 °C	25 °C
Adiabatic efficiency of compressor 85% [16]	85%	85%
Adiabatic efficiency of expander 80% [16]	80%	80%
Minimum temperature approach in heat exchanger (HEX-101, HEX-102, HEX-103)	2 °C	2 °C

Table 2b

Mole fractions of the feed gas for two existing cycles N_2 -CO₂ cycle [18] and N_2 /CH₄-CO₂ [19].

Mole fraction of NG	N_2 -CO ₂ cycle [18]	N_2 /CH ₄ -CO ₂ cycle [19]
CH ₄	0.82	0.90
C ₂ H ₆	0.112	0.05
C ₃ H ₈	0.04	0.02
i-C ₄ H ₁₀	0.012	0.01
n-C ₄ H ₁₀	0.009	0.01
N ₂	0.007	0.01
CO ₂	0	0

compared to that reported in Ref. [18] (Table 5). Furthermore the optimized N_2 /CH₄-CO₂ liquefaction cycle achieved specific energy consumption (SEC) 8.78 kW/kmol/h at liquefaction rate of 0.95, which shows about 3.62% reduction in specific energy consumption (SEC) at same liquefaction rate compared to that reported in Ref. [19] (Table 5).

6.3. Comparison of the modified and optimized N_2 -CO₂ and N_2 /CH₄-CO₂ cycles with similar proposed cycles in literature

6.3.1. Energy analysis

As mentioned before, the results for N_2 liquefaction process with CO₂ pre-cooling (N_2 – CO₂) [18] and mixed refrigerant expansion cycle N_2 /CH₄ process with CO₂ pre-cooling (N_2 /CH₄ – CO₂) [19] as well as the results for the modified and optimized cycles are listed in Table 5.

In both Refs. [18] and [19], expanders were applied for processes with pressure reduction. However, the produced shaft power by expander was used by compressors in compression processes. To simulate N_2 -CO₂ and N_2 /CH₄-CO₂ cycles here, expanders are also used to make the cycles as similar as possible to two above references. Expander power outputs were also used as a part of energy consumption for compressors.

Results showed that the estimated amount of power saved by expanders in N_2 -CO₂ cycle was about 12.43 kW while the power consumption of compressors was about 41.25 kW. Furthermore, in N_2 /CH₄-CO₂ cycle, the expander power output was about 3.2 kW while the power consumption of compressors was about 11.5 kW.

This caused that the modified and optimized N_2 -CO₂ cycle had lower specific energy consumption (SEC) for about 5.25% (by decreasing from $9.90 \frac{kW}{kmol} \cdot h$ to $9.38 \frac{kW}{kmol} \cdot h$) at similar liquefaction rate of 0.77 in comparison with that reported in Ref. [19]. Also the modified and optimized N_2 /CH₄-CO₂ cycle had lower specific energy consumption (SEC) for about 3.62% (by decreasing from $9.11 \frac{kW}{kmol} \cdot h$ to $8.78 \frac{kW}{kmol} \cdot h$) at similar liquefaction rate of 0.95 in comparison with that reported in Ref. [20].

Furthermore, numbers of equipment used in both modified and

Table 3

Comparison of simulation results in this study with that for existing cycles N_2 -CO₂ [18] and N_2 /CH₄-CO₂ [19] for the same input values of Tables 2a and 2b.

Process parameters	Simulated result for N_2 -CO ₂ cycle in this study	Reported results for N_2 -CO ₂ cycle [18]	Difference in percentage points	Simulated result for N_2 /CH ₄ cycle in this study	Reported results for N_2 /CH ₄ cycle [19]	Difference in percentage points
Flow rate of feed gas (kmol/h)	4.0	4.0	0.0%	1.0	1.0	0.0%
Flow rate of refrigerant (kmol/h)	21.6	22.6	4.4%	3.71	3.71	0.0%
Power consumption of compressors (kW)	42.63	42.72	0.2%	11.66	NA	-
Power output of expander (kW)	12.09	12.10	0.1%	3.044	NA	-
Specific energy consumption (kW/kmol/h)	9.915	9.9	0.1%	9.113	9.11	0.0%
Liquefaction rate	0.77	0.77	0.0%	0.95	0.95	0.0%

Table 4a
Operating conditions for the modified and optimized N₂-CO₂ cycle.

Material stream	Temperature (°C)	Pressure (kPa)	Molar flow (kmol/h)	Molar enthalpy (kJ/h)	Molar entropy (kmol/kmol °C)	Energy stream	Heat flow (kJ/h)
101	32	4800	4	−7.82e+004	154.8		
102	−30	4800	4	−8.25e+004	140.1		
103	−30	4800	0.3372	−1.075e+005	106.5		
104	−78	200	0.3372	−1.075e+005	116.4		
105	−40	200	0.3372	−9.942e+004	153.7		
106	30	200	0.3372	−9.2e+004	182.9		
107	−30	4800	3.663	−8.028e+004	143.2		
108	−132.1	4800	3.663	−9.098e+004	88.55		
109	−150.9	200	3.663	−9.095e+004	90.98		
LNG-110	−150.9	200	3.073	−9.347e+004	79.91		
201	32.29	327	20.77	194	139	Q-100	7.289e+004
202	151.4	901	20.77	3703	140.3	Q-101	7.189e+004
203	35	901	20.77	241.7	130.8	Q-102	7.372e+004
204	155.4	2483	20.77	3791	132	Q-103	7.549e+004
205	35	2483	20.77	156.3	122.1	Q-104	4.475e+004
206	−51.9	2483	20.77	−2502	111.9		
207	−134.1	327	20.77	−4656	116.1		
208	−69.40	327	20.77	−2769	127.2		
209	−69.40	327	4.528	−2769	127.2		
210	29.87	327	4.528	123.1	138.8		
211	−69.40	327	16.24	−2769	127.2		
212	32.97	327	16.24	213.8	139.1		
301	32.92	604	0.3	−393,700	158.1	Q-105	977.9
302	118.5	1653	0.3	−390,500	159.4	Q-106	1091
303	35	1653	0.3	−394,100	148.9	Q-107	933.6
304	123	4524	0.3	−391,000	150.1	Q-108	13.99
305	35	4524	0.3	−395,700	136.7		
306	15.9	4524	0.3	−397,100	132		
307	−48.84	604	0.3	−397,100	145.4		
308	−1.85	604	0.3	−395,100	153.2		

optimized N₂ – CO₂ and N₂/CH₄ – CO₂ cycles are compared with those reported in Refs. [19] and [20] in Table 5. Numbers show that modified and optimized N₂ – CO₂ cycle had only one heat exchanger more than that reported in Ref. [18] which had a 14.6% increase in annual investment cost. Also the modified and optimized N₂/CH₄ – CO₂ cycle had the same number of equipment in comparison with that reported in Ref. [20]. Thus almost without adding any equipment, the SEC value of proposed cycles of N₂ – CO₂ and N₂/CH₄ – CO₂ decreased 5.25% and 3.62% respectively.

Comparison of results for the proposed modified and optimized cycles of N₂ – CO₂ and N₂/CH₄ – CO₂ with the similar cycles reported

in literature, showed the low investment cost as well as low SEC values for these proposed cycles. As MR-C3, New MR, N₂-CO₂, N₂-CH₄ cycles have specific energy consumption (SEC) of 7.26, 8.15, 9.90, 17.68 kW/kmole/hr respectively [18], thus SEC values 9.38 and 8.87 which are obtained for the modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ cycles are acceptable values in the given range for SEC values of other similar cycles.

Furthermore, the number of equipment in a cycle with specific amount of feed gas represents the order of magnitude of capital cost of that cycle. For MR-C3, New MRC, N₂-CO₂, N₂-CH₄ cycles, the number of key equipment (including compressor, expander, heat exchanger,

Table 4b
Operating conditions for the modified and optimized N₂/CH₄-CO₂ cycle.

Node	Temperature (°C)	Pressure (kPa)	Molar flow (kmol/h)	Molar enthalpy (kJ/h)	Molar entropy (kmol/kmol °C)	Energy stream	Heat flow (kJ/h)
101	30	5000	1	−7.724e+004	152		
102	−147.6	5000	1	−9.139e+004	81.21		
103	−152.9	200	1	−9.139e+004	82.71		
104-LNG	−152.9	200	0.95	−9.240e+004	79.17		
105	−152.9	200	0.05	−7.334e+004	146.3		
106	−30	200	0.05	−6.785e+004	175.1		
107	30	200	0.05	−6.715e+004	177.5		
201	30	240.3	3.85	−3.282e+004	162.7	Q-100	2.03E+04
202	185.8	1050	3.85	−2.754E+04	164.4	Q-101	2.04E+04
203	32	1050	3.85	−3.285E+04	150.4	Q-102	2.03E+04
204	188.8	4550	3.85	−2.757E+04	152.1	Q-103	2.17E+04
205	32	4550	3.85	−3.321E+04	137.3	Q-104	6720
206	−30	4550	3.85	−3.544E+04	129.1	Q-105	4975
207	−101.8	1050	3.85	−3.718E+04	131.7		
208	−149.7	240.3	3.85	−3.847E+04	134.5		
209	−34.35	240.3	3.85	−3.487E+04	155.1		
301	27.89	1500	0.26	−3.944E+05	148.9	Q-106	505.8
302	83.36	2924	0.26	−3.924E+05	149.7	Q-107	644.5
303	32	2924	0.26	−3.949E+05	142.2	Q-108	474.9
304	89.54	5724	0.26	−3.931E+05	142.9	Q-109	976.5
305	32	5724	0.26	−3.968E+05	131.6		
306	−25.15	1500	0.26	−3.968E+05	139.8		

Table 5

Comparison of liquefaction rate, SEC and the number of equipment for the proposed modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ cycles with those reported by [18] and [19].

Process parameters	Modified and optimized N ₂ – CO ₂	N ₂ – CO ₂ [18]	Modified and optimized N ₂ /CH ₄ – CO ₂	N ₂ /CH ₄ – CO ₂ [19]
Liquefaction rate	0.77	0.77	0.95	0.95
Unit energy consumption ($\frac{kW}{kmol} \cdot h$)	9.38	9.90	8.78	9.11
Num. of compressor(s)	4	4	4	4
Num. of expander(s)	1	1	2	2
Num. of heat exchanger(s)	4	3	2	2
Num. of separator(s)	2	2	1	1
Num. of valve(s)	3	3	2	2

separator and expansion valve) are 24, 13, 12, 10 respectively [18]. The first cycle is the most complex one with the maximum number of equipment while the last cycle is the simplest one with lowest number of equipment. In our study here the modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ cycles had 12 and 11 main equipment respectively which are in the same typical range of number of equipment or investment cost.

6.3.2. Exergy analysis

Table 6 shows the share of exergy destruction of equipment. Results show that the highest value of exergy destruction belongs to compressors and expanders. Table 6 also illustrates that for the modified and optimized N₂-CO₂ cycle, the exergy destruction decreased in main heat exchanger (HEX-101) for about 28% (from 1.44 kW to 1.03 kW). This reduction in exergy destruction was due to selecting optimum design parameters in the cycle which resulted in closer hot and cold temperature curves in HEX-101 as well as recovery of cooling energy from reflux gas. Decrease in exergy destruction of the modified and optimized N₂/CH₄-CO₂ cycle in main heat exchanger (HEX-101) was about 80% (from 0.15 kW to 0.03 kW).

The total exergy destruction of main equipment in two above cycles is shown in Fig. 3. Results show that the total exergy destruction decreased for the modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ cycles for 9% (from 15.25 kW to 13.81 kW) and 16% (from 3.8 kW to 3.19 kW) respectively.

Table 6 and Fig. 3, show that almost one third of the total exergy destruction was for compressors.

6.3.3. Economic analysis

Results of economic analysis for the modified and optimized N₂ – CO₂ and N₂/CH₄ – CO₂ cycles with that reported in [18] and [19] are shown in Figs. 4a and 4b respectively. In economic analysis the system lifetime and interest rate were considered 20 years and 12% respectively.

It was assumed that power generated by expanders was used in compressors. Thus, the operating cost was computed by multiplying the net power consumption (compressor power consumption minus

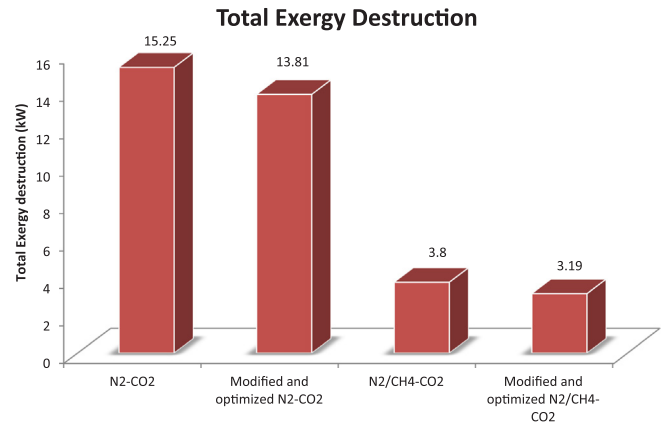


Fig. 3. Comparison of total exergy destruction of main equipment in the modified and optimized N₂-CO₂ and N₂/CH₄-CO₂ cycles with exergy destruction of existing N₂-CO₂ [18] and N₂/CH₄-CO₂ [19] cycles.

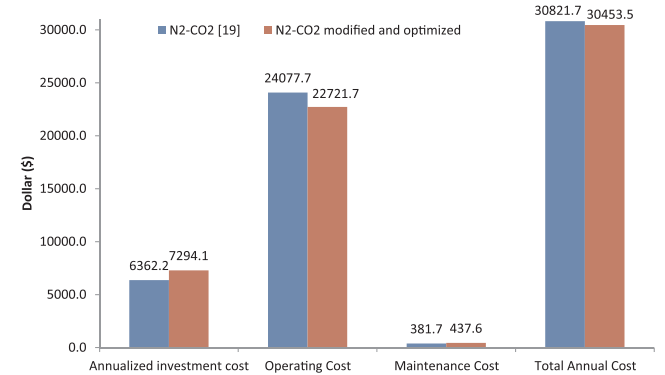


Fig. 4a. Comparison of results for economic analysis for the modified and optimized N₂-CO₂ cycle with that cycle reported in [18].

Table 6

Comparison of results for exergy analysis of equipment for the proposed modified and optimized design of N₂-CO₂ and N₂/CH₄-CO₂ cycles and existing cycles in Refs. [18] and [19].

Cycle	Components				
	Exergy destruction	Main Heat exchanger (HEX101)	Compressors	Expander(s)	Valves
Modified and optimized N ₂ – CO ₂	Value (kW)	1.03	4.37	7.14	1.27
	Percentage (%)	7.5	31.6	51.7	9.2
N ₂ – CO ₂ [18]	Value (kW)	1.44	4.63	7.11	2.07
	Percentage (%)	9.4	30.4	46.6	13.6
Modified and optimized N ₂ /CH ₄ – CO ₂	Value (kW)	0.03	1.15	1.71	0.30
	Percentage (%)	0.9	36.1	53.6	9.4
N ₂ /CH ₄ – CO ₂ [20]	Value (kW)	0.15	1.19	1.60	0.86
	Percentage (%)	3.9	31.3	42.1	22.6

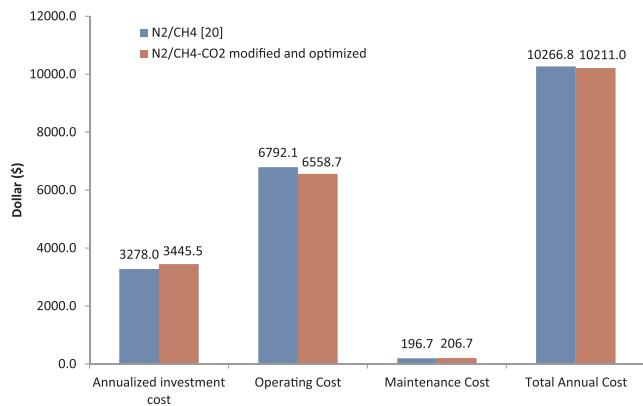


Fig. 4b. Comparison of results for economic analysis for the modified and optimized N_2/CH_4-CO_2 cycle with that cycle reported in [19].

expander power production) by working hours in a year and by 9 cents per kW h for buying electricity [31].

For the modified and optimized $N_2 - CO_2$ cycle, 14.6% increase (from 6362.2\$ to 7294.1\$) in annual investment cost in comparison with that for the cycle discussed in [18] was observed. However, due to lower operating cost of this cycle (from 24077.7\$ to 22721.7\$) for about 5.6%, the total annual cost decreased (from 30821.7\$ to 30453.5\$) for about 1.19%.

For $N_2/CH_4 - CO_2$ modified and optimized cycle, the operational cost decreases (from 6792.1\$ to 6558.7\$) for about 3.4% while the annual investment cost increased (from 3278\$ to 3445.5\$) for about 5.1%, resulting decrease in total annual cost (from 10266.8\$ to 10211\$) for about 0.5%. The main reason for increasing the annual investment cost was selecting bigger heat exchangers (heat transfer surface area) with lower hot and cold temperature differences.

6.4. Performance analysis of the modified and optimized processes

To analyze the output results for the modified and optimized N_2-CO_2 and N_2/CH_4-CO_2 cycles at various feed gas pressure and temperature, the values of power consumption and liquefaction rate were estimated for the above various conditions. The performance of both proposed cycles after modification and optimization under different feed gas conditions are illustrated in Figs. 5a and 5b and Figs. 6a and 6b. These results are discussed in the following sections.

6.4.1. Effect of feed gas pressure

Feed gas pressure has an important impact on SEC and liquefaction rate values for both N_2-CO_2 and N_2/CH_4-CO_2 cycles (Figs. 5a and 5b).

With increasing the feed gas pressure in N_2-CO_2 cycle and with more heavy hydrocarbons in liquid phase in separator (S-100), thus lower amount of natural gas (NG) flow rate (q-107), enters HEX-103. For cooling this lower amount of NG (as hot fluid) and for its phase change, lower refrigerant (as cold fluid) molar flow rate and compressor power input were required. Thus with higher feed gas pressure, lower NG molar flow rate, lower liquefied NG molar flow rate and from all above, lower SEC value were obtained.

With increasing the feed gas pressure in N_2/CH_4-CO_2 cycle, no significant change in NG liquefaction was observed due to the fact that temperature and pressure after expansion valve VLV-101 did not change. Furthermore, with increasing the feed gas pressure, the refrigeration effect could be gained by expansion valve VLV-101 with lower amount of refrigerant molar flow rate which decreases SEC value.

6.4.2. Effect of feed gas temperature

The next controlling parameter in LNG production is the feed gas temperature. Liquefaction rate and SEC value are shown for N_2-CO_2 and N_2/CH_4-CO_2 cycles in Figs. 6a and 6b.

With increasing the feed gas temperature in N_2-CO_2 cycle, more cooling effect and more refrigerant molar flow rate are required in HEX-102 (node-102) heat exchanger. This increases the compressor power consumption. As the temperature and pressure after HEX-102 do not change significantly, thus with almost no change in liquefaction rate, SEC value increases.

The same above expressions could be used for N_2/CH_4-CO_2 cycle with HEX-101 (node-103).

7. Conclusions

Two improved and optimized cycles were proposed for small-scale natural gas liquefaction. The first cycle was nitrogen expansion with carbon dioxide pre-cooling (N_2-CO_2) and the second cycle was mixed refrigerant nitrogen and methane expansion cycle with carbon dioxide pre-cooling (N_2/CH_4-CO_2).

Two above cycles, were selected from Refs. [18] and [19]. Then both above cycles were first modified by new arrangement of equipment or adding equipment such as new heat exchangers or distributors. The composite temperature curves became closer in HEX-101 after the above modifications.

Then both cycles were optimized by selecting optimum values for operating parameters (design parameters or decision variables) and compared with those values reported in [18] and [19]. Twelve design parameters for N_2-CO_2 and thirteen design parameters for N_2/CH_4-CO_2 were specified. With these optimum design parameters, energy analysis showed that the specific energy consumption to produce the same liquefaction molar flow rate for N_2-CO_2 and N_2/CH_4-CO_2 cycles dropped for 5.25% (from 9.90 to 9.38 kWh/kmole) and 3.62% (from 9.11 to 8.78 kWh/kmole) in comparison with those values reported in [18] and [19].

Exergy analysis also illustrated 9.4% (from 15.25 to 13.81 kW) and 16% (from 3.8 to 3.19 kW) drop in exergy destruction in the modified and optimized N_2-CO_2 and N_2/CH_4-CO_2 cycles in comparison with those for Refs. [18] and [19]. This lower exergy destruction was mainly due to lower difference values of composite temperature curve in HEX-101. This closer temperature curves provided 28% (from 1.44 to 1.03 kW) and 80% (from 0.15 to 0.03 kW) drop in exergy destructions in N_2-CO_2 and N_2/CH_4-CO_2 cycles in comparison with those in Refs. [18] and [19].

Besides energy and exergy analysis, the economic analysis also showed 1.19% (from 30821.7 to 30453.5 dollars) and 0.5% (from 10266.8 to 10,211 dollars) drop in total annual cost (sum of investment, maintenance and operational costs) for N_2-CO_2 and N_2/CH_4-CO_2 cycles in comparison with those in Refs. [18] and [19].

Furthermore, the modified and optimized cycles are as simple as those in [18] and [19] with same compactness and proper operation.

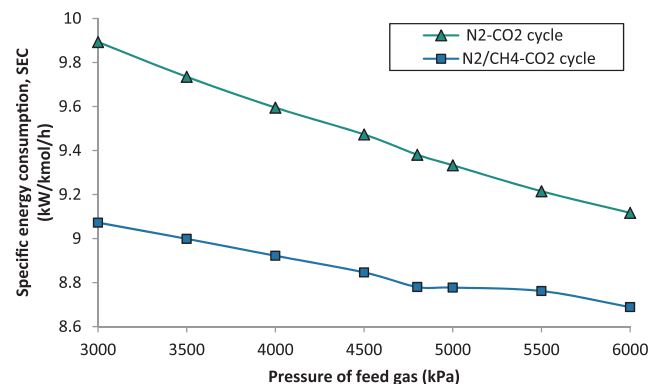


Fig. 5a. Variations of optimum value of specific energy consumption per molar flow rate of LNG production versus feed gas pressure for modified and optimized N_2-CO_2 and N_2/CH_4-CO_2 cycles.

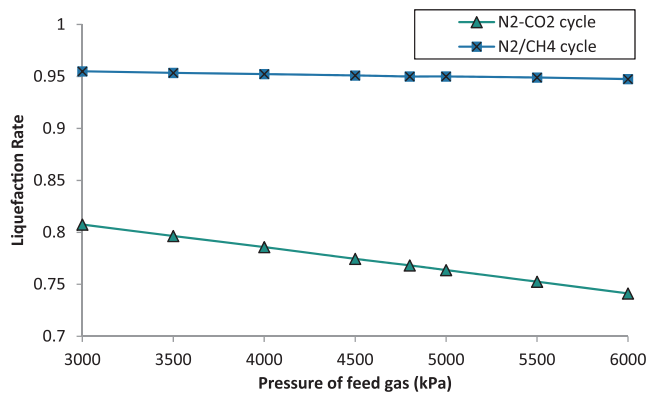


Fig. 5b. Variations of optimum value of liquefaction rate versus feed gas pressure for modified and optimized N₂–CO₂ and N₂/CH₄–CO₂ cycles.

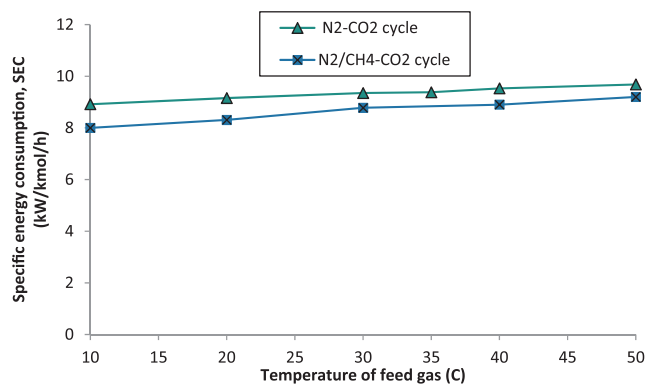


Fig. 6a. Variations of optimum value of specific consumption of LNG production versus feed gas temperature for modified and optimized N₂–CO₂ and N₂/CH₄–CO₂ cycles.

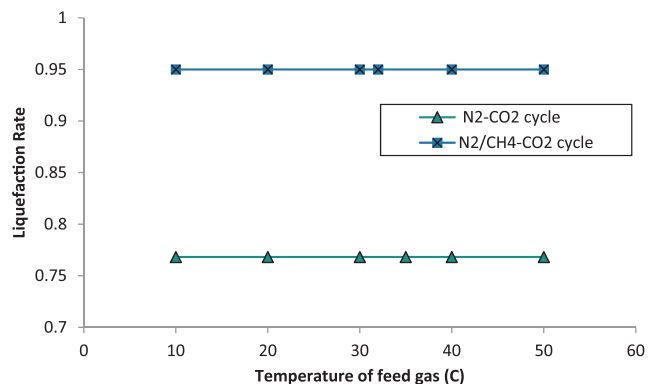


Fig. 6b. Variations of optimum value of liquefaction rate versus feed gas temperature for modified and optimized N₂–CO₂ and N₂/CH₄–CO₂ cycles.

The operation of two cycles were checked for various feed gas pressures and temperatures. Furthermore, the modified and optimized cycles are as simple as those in [18] and [19] with the same compactness and proper operation. The operation of two cycles were checked for various feed gas pressures and temperatures.

Declaration of Competing Interest

There is no conflict of interest for authors of this submitted manuscript.

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