

Exergy and Energy Analysis of Propane Precooled Mixed Refrigerant Process for Liquefaction of Natural Gas

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Alireza Rasekh

Abstract – In this work, liquefied natural gas (LNG) production cycle by means of Propane Precooled Mixed Refrigerant (PPMR) Process has been studied. Energy and exergy steady equations of equipments in the PPMR cycle have been established. The equipments are described using rigorous thermodynamics and no significant simplification is assumed. Taken some operating parameters as key parameters, influences of these parameters on coefficient of performance (COP) and exergy efficiency of the cascading cycle are analyzed. The results indicate that the PPMR cycle has good performance, with COP and exergy efficiency of 1.725 and 37.78%, respectively, for a typical operating condition. The power consumed for liquefaction of natural gas (NG) is equal to 42.2 MW. Parametric analyses are performed for the PPMR cycle to evaluate the effects of key factors on the performance of this process through simulation calculations. Results show that the COP and exergy efficiency will be improved with increasing of the inlet pressure of mixed refrigerant (MR) compressors, decreasing of the NG and MR temperature after precooling process, outlet pressure of turbine, inlet temperature of MR compressor and NG temperature after cooling in Mean Cryogenic Heat Exchanger (MCHE).

Keywords - Coefficient of performance, Energy efficiency, Exergy efficiency, LNG, PPMR process.

1. INTRODUCTION

Natural gas is often found in remote locations far from developed industrial nations. For the transportation of NG from producing wells to utilization sites, two approaches are now applied with their respective pros and cons [1-4]. Where possible, the gas is transported by pipeline to the end user. Currently, when oceans separate the gas source and the user, the only viable way to transport the gas is to convert it into LNG and convey it using insulated LNG tankers. LNG is regarded as a relatively clean energy resource. During the process of its preparation, approximately 500 KWh energy/t LNG is consumed for compression and refrigeration and a considerable portion of this invested exergy is preserved in the LNG [5], which has a final temperature of about 110 K, much lower than that of the ambient or of seawater. The liquefaction reduces its volume 600-fold and thus makes long distance transportation convenient. With the increasing demand for cleaner fuels, LNG is now playing an even significant role as energy resource.

The Propane Precooled Mixed Refrigerant process consists of two cycle process for liquefaction of NG using propane in precooled and mixed refrigerant in main cooling. This process currently holds 88% of the liquefaction plants on the market in which produce 107.5 MTPA of LNG with 53 trains in operation. It became the dominant liquefaction process technology by the late 1970s and continues to be the workhorse of the LNG industry today [6].

In previous studies, Barclay *et al.* [7] presented an excellent review on the selection of thermodynamic refrigeration cycle for distributed NG liquefaction.

¹Corresponding author; Tel, Fax: +98 1212223669. E-mail: <u>alireza.rasekh@gmail.com</u>. Joule-Thomson cycle with MR is recommended to take advantages of lower capital costs, as it employs a throttle valve (isenthalpic expansion) with two-phase refrigerant. Another possibility is Claude or Heylandt cycle that combines the isenthalpic and isentropic expansion [8]. Andress and Watkins [9] described their optimized cascade LNG process with highlighting the advantages of safe and easy operation. Chang et al. [10] investigated on methane liquefaction system by Reversed-Brayton cycle thermodynamically and economically. Kanoglu [11] provides an exergy analysis of the multistage cascade refrigeration cycle used for LNG production. He indicated that the minimum work depends only on the properties of the incoming and outgoing NG, and it increases with decreasing liquefaction temperature. Foerg et al. [12] performed the energy analysis of different LNG processes, which suggested that the most efficient LNG processes overall, is the PPMR process. Different refrigeration cycles with different refrigerants can be used for NG liquefaction [13-14]. A common approach in these references is to consider LNG as a substance defined simply by methane properties.

In this work, propane precooled mixed refrigerant process for LNG production has been studied thermodynamically. Exergy and Energy efficiency are important for LNG production as feed gas is consumed in order to carry out the liquefaction process. The influences of important parameters on increasing efficiency of LNG production cycle through PPMR process have been investigated and main results are described.

^{*}Department of Mechanical Engineering, Shahrood University of Technology, Shahrood, Iran.

2. THE DESCRIPTION OF PPMR PROCESS

The liquefaction process of LNG is represented in Figure 1. This process accounts for a very significant proportion of the world's baseload LNG production capacity. There are two main stages for cooling and liquefying NG in PPMR process. At first, NG is cooled in precooling cycle to -40 \circ C [6]. The precooling cycle uses a pure component, propane. The liquefaction and

subcooling cycle uses MR made up of nitrogen, methane, ethane and propane. The NG and MR composition are shown in Table. 1. The LNG production cycle includes two parts. The left hand side in Figure 1 is pertaining to precooling of NG by propane as refrigerant and the right hand side in Figure 1 is subcooling by MR [6]. The features of precooling and subcooling of NG liquefaction are described in the following sections.

Table 1. Mass composition for LNG and MR.

Component -	Composition (mass %)		
- Component	LNG	MR	
Nitrogen (N2)	5	7	
Methane (C1)	87	38	
Ethane (C2)	6	41	
Propane or heavier (C3+)	2	14	



Propane Precooling Cycle

The precooling cycle uses propane at five pressure levels and can cool the process NG down to $-40 \circ C$ [6]. It is also used to cool and partially liquefies the MR. The corresponding T–s diagram is shown in Figure 2. It is mainly composed of eight heat exchangers, four compressors and a condenser. The main cooling fluid is propane. The propane precooling cycle can be identified as M-A-B-D-E-G-H-I-V-K-J-F-C which NG and MR are cooled as 1-2 and 15-16 respectively (Figure 1). The cooling is achieved in kettle-type exchangers with propane refrigerant boiling and evaporating in a pool on the shell side, and with the process streams flowing in immersed tube passes. As to propane precooling, propane with high pressure (20 bar) enters condenser 1 that its temperature is fallen ($30 \circ C$), and next propane is expanded to the lower pressure (8.7 bar) in the throttle valve [6]. Then it goes through a heat addition process in the heat exchanger (HE1,.., HE8) in four stages, the liquefied propane is entranced to next heat exchanger and evaporated propane is moved to compressor that can thereby produce refrigeration which cool the NG and MR down to -40 °C and -30°C respectively. Four centrifugal compressors with side streams recover the evaporated propane streams and compress the vapor to 20 bar to be condensed against water and recycled to the propane kettles [6].

Subcooling Cycle by MR

The subcooling cycle uses MR to liquefy NG and can cool NG to -140 °C [6]. The corresponding T-s diagram is shown in Figure 3. It is mainly composed of MCHE and three compressors and condensers. The subcooling cycle can be identified briefly as 9-10-11-...-25-26 and NG is cooled as 2-3. In the MR cycle, the partially chilled refrigerant that is cooled to -35 °C by propane is separated into two streams (17, 24) that are used to liquefy and subcooling the process stream from typically -35 °C to -150 °C [6]. This is carried out in a proprietary spiral wound exchanger, the MCHE. The MCHE consists of two or three tube bundles arranged in a vertical shell, with the process NG and refrigerants enter the tubes at the bottom which then flow upward under pressure. The process NG passes through all the bundles to emerge liquefied at the top. The liquid MR stream is extracted after the warm or middle bundle and is flashed across a Joule Thomson valve or hydraulic expander onto the shell side. It flows downwards and evaporates,

providing the bulk of cooling for the lower bundles. The vapor MR stream passes to the top (cold bundle) and is liquefied and subcooled, and is flashed across a JT valve into the shell side over the top of the cold bundle. It flows downwards to provide the cooling duty for the top bundle and part of the duty for the lower bundles. The overall vaporized MR stream (9) from the bottom of the MCHE is recovered (-41°C, 4.4 bar) and compressed by the MR compressors (C1, C2, C3) to 52 bar [6]. After that, It is cooled first by water and then by the propane refrigerant, and recycled to the MCHE.

As to LNG production process, two stages are implemented for NG cooling. The corresponding T-s diagram for LNG production is depicted in Figure 4. In the propane precooling, NG temperature is reduced from $35 \circ \text{C}$ to -40 $\circ \text{C}$. Afterward NG is further cooled in MR cycle as the temperature is reached to near -140 $\circ \text{C}$. The high pressure NG is expanded in turbine T1 in which NG temperature is more lowered to -162 $\circ \text{C}$ in almost atmospheric pressure. Therefore whole NG is liquefied and LNG is capable to use. In PPMR cycle, some of produced LNG compressed by compressor C4 to yield high pressure (HP) fuel gas.





Fig. 3. T-s diagram for MR subcooling cycle.



Fig. 4. T-s diagram for LNG production.

3. THERMODYNAMIC ANALYSIS

In order to analyze of the system performance, the influence of several key parameters such as NG and MR temperature after precooling process, outlet pressure of turbine T1 of open LNG cycle, inlet temperature of compressor C3, NG temperature after cooling in MCHE and inlet pressure of MR compressors C1, C3 on coefficient of performance of system and exergy efficiency of the PPMR cycle were evaluated. Thermodynamic analysis is based on a careful numerical simulation which has been performed with the commercial program Aspen HYSYS[®] [15]. To determine the performance of the indicated system, the steady-state component models are used. Every component is modeled in consideration of mass, energy

and species balances. Energy steady equations and exergy steady equations of each unit for the cascading cryogenic cycle are established with neglect of pressure drop and heat loss in heat exchangers and pipelines. The most relevant assumptions for the calculations in this paper are summarized in Table 2.

Energetic and Exergetic Efficiency

The Liquefaction facility is a cryogenic system, which has the aim to transfer heat from cold source to hot by consuming electric power. It can be defined a process efficiency based on the first thermodynamic principle (energetic efficiency) and a process efficiency based on the second thermodynamic principle (exergetic efficiency) by two different point of view.

Table 2. Main assumptions for the calculation.

Parameter	value
inlet NG source temperature (°C)	35
inlet NG source pressure (bar)	45
NG/LNG mass flow rate (Kg/s)	50
Isentropic efficiency of turbine	0.8
Isentropic efficiency of compressor	0.8
LNG pressure drop in precooling (bar)	5
LNG pressure drop in MCHE (bar)	2
Ambient temperature (°C)	20
Ambient pressure (bar)	1

Energy Balance Equations and Energetic Efficiency

For compressor C1, C2, C3, C4 and C5, the energy equation is:

$$W_{j} = m_{j,inlet}(h_{j,outlet} - h_{j,inlet}) = \frac{m_{j,inlet}(h_{j,outlet} - h_{j,inlet})}{\eta_{c}}$$
(1)
$$i = CLC2, C3, C4, C5$$

For turbine T1, the energy equation is:

$$W_i = m_{i,inlet} (h_{i,inlet} - h_{i,outlet}) = \eta_T m_{i,inlet} (h_{i,inlet} - h'_{i,outlet})$$

$$i = T1$$

(2)

For heat exchanger HE1, HE2,..., HE9, the energy equation is:

$$m_c(h_{c,outlet} - h_{c,inlet}) = m_h(h_{h,inlet} - h_{h,outlet})$$
(3)

For MCHE, the energy equation is:

$$m_{LNG}h_2 + m_{22}h_{22} + m_{18}h_{18} + m_{26}h_{26} + m_{21}h_{21} = m_{LNG}h_3 + m_{25}h_{25} + m_{23}h_{23} + m_{9}h_{9}$$
(4)

The whole facility can be considered as a cryogenic system producing LNG. Then the input energy is represented by net work is exerted to system for LNG production. Compressor C4 does not interfere in LNG production and just used for HP fuel gas; due to this fact, the work consumed by compressor C4 is neglected in calculation. After that, the output energy is heat supplied by refrigerants in stages of cooling NG. This approach allows defining the first thermodynamic principle energetic efficiency with the relationship (coefficient of performance of system is usually used in cryogenic application).

A. Rasekh / International Energy Journal 12 (2011) 95-104

$$COP = \frac{m_{LNG}(h_1 - h_3) + m_{MR}(h_{15} - h_{16})}{\Sigma W_j - \Sigma W_i}$$
(5)

$$j = C1, C2, C3, C5; \ i = TI$$

Exergy Balance Equation and Exergetic Efficiency

For the fluid of unit mass, the exergy is defined as $e = (h - h_0) - T_0(s - s_0)$ (6)

Overall exergy balance equation is

$$E_{in} = E_{eff} + E_{loss} \tag{7}$$

The effective exergy of the cycle is the heat provided in NG cooling equals to the sum of the refrigeration exergy produced in the evaporators in precooling and subcooling.

$$E_{eff} = E_{precooling} + E_{subcooling} \tag{8}$$

The exergy produced in the evaporators in precooling can be obtained as,

$$E_{precooling} = m_{LNG} \left(e_2 - e_1 \right) + m_{MR} \left(e_{16} - e_{15} \right) \tag{9}$$

The exergy produced in subcooling equals to,

$$E_{subcooling} = m_{LNG} \left(e_3 - e_2 \right) \tag{10}$$

And the both the NG exergy and net given work to system is considered as the inputs.

$$E_{in} = \sum E_j - \sum E_i + E_{NG} \quad j = C1, C2, C3, C5; \ i = T1$$
(11)

Table 3. The stream parameters of the PPMR cycle.	
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The exergy E_i for turbine T1 in Eq. (11) is:

$$E_i = m_{i,inlet} (e_{i,inlet} - e_{i,outlet}); \quad i = TI$$
(12)

The consumed exergy E_i for compressor in Eq. (11) is:

$$E_{j} = m_{j,inlet} (e_{j,outlet} - e_{j,inlet}); \quad j = C1, C2, C3, C5$$
(13)

The exergy efficiency of the cycle is:

$$\eta_{ex} = \frac{E_{eff}}{E_{in}} \tag{14}$$

Table 3 summarizes the thermodynamic properties including temperature, pressure, enthalpy, entropy, exergy, mass flow rate and vapor fraction of each stream for the PPMR cycle where parameters in table. 2 remain constant. With 50 kg/s mass flow rate of NG at the inlet of cycle taken as assumption according to Table 1, the mass flow rates of propane in precooling and MR in subcooling can be determined.

Performance of the described PPMR process under the typical operating condition is presented in Table 4. This system can consume a net power of about 42 MW while the net exergy efficiency is estimated about 37.78%.

Stream no.	T(∘C)	P(bar)	h (kJ/kg)	s (kJ/kg K)	e (kJ/kg)	m(kg/s)	Х
1	35	45	-19.8	-1.994	566.9	50	1
2	-40	40	-208.4	-2.643	568.2	50	1
3	-140	38	-829.9	-6.088	956.1	50	1
4	-161.6	1	-842.2	-6.06	935.8	50	0.13
5	-161.6	1	-911	-6.667	1049	44.607	0
6	-161.6	1	-400.1	-2.089	218.1	5.393	1
7	-40	1	-142.2	-0.5335	17.07	5.393	1
8	250.7	25	575.1	-0.2489	646.7	5.393	1
9	-41	4.4	-149.5	-1.329	242.1	159.3	1
10	51.2	14.1	47.4	-1.204	402.6	159.3	1
11	30.2	14.1	-1.9	-1.91	560.1	159.3	1
12	46.3	45	8.6	-1.903	568.8	159.3	1
13	30	45	-32.4	-2.036	566.3	159.3	1
14	43.14	52	-5.5	-2.019	588.2	159.3	1
15	30	52	-39.4	-2.128	586.4	159.3	1
16	-35	52	-217.2	-2.79	602.7	159.3	1
17	-35	52	-217.2	-2.79	602.7	19.91	1
18	-68.1	8.9	-217.2	-1.987	367.3	139.4	1
19	-70	52	-357	-3.431	650.6	9.955	1
20	-145	4.4	-357	-2.478	371.3	9.955	1
21	-145	4.4	-357	-2.478	371.3	19.91	1
22	-68.1	8.9	-217.2	-1.987	367.3	9.955	1
23	-126.3	8.9	-357	-2.789	463.7	9.955	1
24	-35	52	-217.2	-2.79	602.7	139.4	1
25	-76	8.9	-221.3	-2.007	369	139.4	1
26	-145	4.4	-357	-2.478	371.3	139.4	1

Table 4. Cycle performance summary.	
power output by Turbine T1, MW	0.609
Power consumed by compressor C1, MW	31.3
Power consumed by compressor C2, MW	1.68
Power consumed by compressor C3, MW	4.28
Power consumed by compressor C4, MW	3.84
Power consumed by compressor C5, MW	1.62
Net power input, MW	42.2
Total thermal power supplied in precooling stage from NG, MW	9.43
Total thermal power supplied in precooling stage from MR, MW	28.31
Total thermal power supplied in subcooling stage from NG, MW	31.07
Propane mass flow rate, kg/s	20
MR mass flow rate, kg/s	159.3
Coefficient of performance (COP)	1.725
Exergy efficiency	37.78%

4. RESULTS AND DISCUSSIONS

In order to investigate the effect of the parameters on the coefficient of performance and exergy efficiency of the considered cycle, analysis has been conducted. According to energy steady equations and exergy steady equations of the PPMR cycle, taken NG and MR temperature after precooling by propane, outlet pressure of turbine T1, NG temperature after cooling in MCHE in open LNG cycle, inlet pressure of MR compressor C1 and inlet temperature and pressure of compressor C3 as key parameters, influences of these parameters on COP and exergetic efficiency of the PPMR cycle has been analyzed. The values were obtained by using of the commercial program Aspen HYSYS®, in which the component models are based on the energy balance and mass balance, with the default relative convergence error tolerance of 0.0001% which is used to determine whether a tear stream is converged or not, the tear stream is one for which Aspen HYSYS makes an initial guess, and iteratively updates the guess until two consecutive guesses are within a specified tolerance. The tear stream is converged when the following is true for all tear convergence variables X including the total mass flow, all component mass flows, pressure, and enthalpy:

 $-tolerance < [(X_{calculated} - X_{assumed})/X_{assumed}] < tolerance$

Where the default for tolerance is 0.0001, $X_{assumed}$ is the assumed value of X before the calculation is conducted; $X_{calculated}$ is the calculated value of X.

The Peng-Robinson equation of state is used for thermodynamic calculations [16]. It is a well known equation of state for calculation of light hydrocarbons and pure components. The amount of pressure and temperature of selected parameters are constrained between actual operation conditions.

Figure 5 shows the effect of the NG temperature after precooling process on the COP and exergy efficiency where NG is cooled by propane at pressure of 45 bar and the rest conditions are listed in Table 3. It is seen that both the COP and exergy efficiency of the cycle are decreasing with the NG temperature after precooling process. However NG temperature increasing after precooling brings about needed power become lower for propane compresses (C5_a, C5_b, C5_c and C5_d), but it was induced supplied heat in heat exchangers

(HE1, ..., HE4) from NG to propane at precooling process is decreased, and so both the COP and exergy efficiency will be declined.

The impact of the MR outlet temperature from precooling process on the COP and exergy efficiency is presented in Figure 6. It can be seen that both the COP and exergy efficiency is decreasing with increasing MR temperature after precooling by propane. Like NG temperature, MR temperature increasing after precooling is caused captured heat by propane in heat exchangers at precooling process is lessened, therefore both the COP and exergy efficiency will be diminished.

Changes of COP and exergy efficiency of the PPMR process with outlet pressure of turbine T1 are shown in Figure 7, where the inlet pressure of the turbine T1 is 38 bar. We can see that both the COP and exergy efficiency of the PPMR cycle decrease with the increasing of outlet pressure of turbine T1 because higher outlet pressure of turbine T1 is, lower output of power of turbine T1 is.

Figure 8 describes the variations of COP and exergy efficiency of this cryogenic cycle with NG outlet temperature from MCHE in open LNG cycle where heat transfer is occurred under pressure of 38 bar for NG. It is seen from Figure 8, the COP and exergy efficiency will decrease with NG outlet temperature from MCHE. Although increasing NG outlet temperature from MCHE is induced required input power for MR compressors (C1, C2 and C3) is reduced; but total captured heat from NG in MCHE will be also decreased and eventually COP and exergy efficiency will be decreased.

The influence of inlet temperature of compressor C3 on the COP and exergy efficiency is shown in Figure 9, where the inlet pressure of the compressor C3 is 45 bar and the rest conditions are listed in Table 3. As seen, both the COP and exergy efficiency of the cryogenic cycle are decreasing when the inlet temperature of compressor C3 goes higher. The changing tendency of the curves is similar to each other. This is because the input power to compressor C3 increases with the increasing of the inlet temperature of compressor C3. Hence, the COP and exergy efficiency of the cycle will be decreased. In addition, Figure 9 also shows that even the inlet temperature of is more, the COP and exergy efficiency can also reach about 1.783 and 37.62%, respectively.



Fig. 5. The effect of the NG temperature after precooling process on the COP and exergy efficiency.



Fig. 6. The effect of the MR temperature after precooling process on the COP and exergy efficiency.



Fig. 7. The effect of the outlet pressure of turbine T1 on the COP and exergy efficiency.



Fig. 8. The effect of NG outlet temperature from MCHE in open LNG cycle on the COP and exergy efficiency.

The effect of the inlet pressure of MR compressor C1 on the COP and exergy efficiency is shown in Figure 10. It is seen both the COP and exergy efficiency are increasing with MR pressure. More MR inlet pressure of compressor brings about required input work to MR compressor is decreased while supplied heat to MR is almost remaining constant. Hence both the COP and exergy efficiency will be improved with inlet pressure of MR compressor C1.

Figure 11 shows the effect of inlet pressure (P_{13}) of MR compressor C3 on the COP and exergy efficiency. From Figure 11, it can be realized both the COP and exergy efficiency are intensifying with inlet pressure of MR compressors. Like inlet pressure of MR compressor C1, increasing inlet pressure of MR compressor C3 is caused required power to compressor is decreased and therefore COP and exergy efficiency of PPMR cycle

will be increased.

Figure 12 shows the variations of COP with NG liquefaction temperature for multistage cascade refrigeration (MCR) and PPMR process. However these two processes aren't identical but there is slightly distinction between them. For this reason, MCR process can be considered as a reliable criterion for calculations. A comparison between the calculated values in PPMR process and the MCR process is given in Figure 12. As the figure shows, the results obtained with the calculations are in good agreement when compared with those obtained in the literature [11]. Furthermore, figure 12 indicates that the maximum possible liquefaction temperature should be used to maximize the COP. In other words, the LNG should not be liquefied to the lower temperatures than needed.



Fig. 9. The effect of inlet temperature of compressor C3 on the COP and exergy efficiency.



Fig. 10. The effect of inlet pressure of MR compressor C1 on the COP and exergy efficiency.



Fig. 11. The effect of inlet pressure of MR compressor C3 on the COP and exergy efficiency.



Fig. 12. Variations of COP with NG liquefaction temperature for MCR and PPMR process.

5. CONCLUSION

The PPMR process for liquefied NG production has been evaluated and thermodynamically modeled. Energy steady equations and exergy steady equations of heat exchangers and power equipments in the cascading cryogenic cycle were established. The equipments were described using rigorous thermodynamics and no significant simplification was assumed. Heat duty of each component was indicated and power of each compressor was estimated, COP and exergy efficiency of this cycle were calculated.

In order to increase the COP and exergy efficiency of the cryogenic cycle, some key parameters have been analyzed. The results show that the NG and MR temperature after precooling process and inlet pressure of MR compressors C1 and C3 affect the COP and exergy efficiency of the LNG production cycle.

When the NG and MR temperature after precooling decrease or inlet pressure of MR compressors C1 and C3 increase both the COP and exergy efficiency will be improved. The outlet pressure of turbine T1, NG temperature after cooling in MCHE and inlet temperature of compressor C3 are other factors affecting the efficiency of the LNG production cycle. The COP and exergy efficiency will increase with decreasing of outlet pressure of turbine T1, NG temperature after cooling in MCHE and inlet temperature after cooling in MCHE and inlet temperature after cooling in MCHE and inlet temperature of compressor C3. The exergy efficiency and COP of the PPMR process cycle are calculated as 1.725 and 37.78%, respectively, while the power consumed for liquefaction of NG is equal to 42.2 MW for a typical operating condition.

Calculated values in PPMR are compared to the MCR ones in which the two cycle are mainly similar to each other. The agreement between calculated and those obtained in the literature is mostly good.

NOMENCLATURE

e	specific exergy (kJ/kg)
Е	exergy (kW)
h	specific enthalpy (kJ/ kg)
m	mass flow rate (kg/s)
Р	pressure (bar)
Q	rate of heat flow (MW)
S	specific entropy (kJ/kg K)
Т	temperature (°C)

W	work (MW)
Х	Vapor fraction
Abbreviations	
С	compressor
COP	coefficient of performance
HE	heat exchanger
LNG	liquefied natural gas
MCHE	mean cryogenic heat exchanger
Greek symbols	
η_{ex}	exergy efficiency
η_{T}	isentropic turbine efficiency
η_c	isentropic compressor efficiency
Subscripts	
с	cold stream
ex	exergy
ef	efficient
h	hot stream
1-26	states on the cycle flowsheet
A-M	states on the cycle flowsheet
0	reference state
T1	LNG turbine
in	input

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104

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A. Rasekh / International Energy Journal 12 (2011) 95-104

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