

Energy Integration of LNG Light Hydrocarbon Recovery and Air Separation

Ruihang Zhang^a, Chufan Wu^b, Wuwenjie Song^b, Chun Deng^{a,*}

^aState Key Laboratory of Heavy Oil Processing, College of Chemical Engineering and Environment, China University of Petroleum, Beijing, 102249, China

^bSchool of Chemical Engineering and Analytical Science, The University of Manchester, Manchester M13 9PL, United Kingdom

chundeng@cup.edu.cn

Liquefied natural gas (LNG) needs to be heated and vaporized before entering the natural gas pipeline network. The traditional seawater heating method not only wastes the cold energy from LNG regasification, but also causes adverse environmental impact, hence the design of cold energy utilization system is necessary. Air separation process is the most common type used in Chinese LNG receiving terminals, and the utilization ratio of LNG cold energy is only about 40 %. The light hydrocarbon separation process is another way to utilize cold energy, which can recover high-value hydrocarbons like ethane from LNG. This paper first presents an integrated process of light hydrocarbon separation and air separation to realize the cascade utilization of LNG cold energy and obtain higher economic benefits. Then, different temperature ranges of LNG regasification for the air separation and light hydrocarbon separation are determined by sensitivity analyses. Results show the utilization ratio of LNG cold energy is increased by 35.77 %. Air separation and light hydrocarbon separation can be easily miniaturized, and they are suitably applied for the LNG receiving terminals.

1. Introduction

In recent years, China has been increasingly imported LNG with a growth rate of 48% in 2017, exceeding the South Korea becomes the second largest LNG importer in the world. Correspondingly, lots of LNG receiving terminals are planned to be built in Chinese coastal areas. At present, more than 60 known projects have been completed and 18 of them have already putted into production. Among these LNG receiving terminals, the air separation is the most commonly used cold energy utilization system (Xu et al., 2013).

Air separation is a process of extracting liquid nitrogen and liquid oxygen from air by cryogenic method. The cryogenic state is usually achieved through multiple compression, water cooling and throttling, which consuming a great amount energy. Therefore, LNG used as a refrigerant can save this energy. In 1979, Yamanouchi and Nagasawa (1979) proposed a method of using LNG cold energy for air separation. After that, many more in-depth studies have been conducted. Morosuk et al. (2014) designed an energy integration process of air separation and LNG re-vaporization, which proved that using LNG had lower energy consumption and higher product quality. Tesch et al. (2016) carried out conventional and advanced exergy analyses for this integration process. Ebrahimi and Ziabasharhagh (2017) conducted thermodynamic and economic analyses for this integration process. Kim et al. (2018) analyzed and compared two methods of using cold energy of LNG. The LNG stream was used as an extra refrigeration source in a liquid nitrogen production cycle or for precooling of air and nitrogen streams. In China, Xiong and Hua (2007) compressed circulating nitrogen twice and produced three streams in a LNG heat exchanger to receive the cold energy of LNG. Fan et al. (2007) designed a LNG heat exchanger, using LNG to cool both circulating nitrogen and air entering the device. Xu et al. (2014) simplified air separation process and widened the operating temperature range of LNG by high-efficiency heat exchanger network and chemical packing separation technology. Recently, Chen et al. (2019) studied the distillation column which is most suitable for the integrated process of air separation and LNG regasification. These processes are developed to achieve the goal of cold energy recovery and production of liquid nitrogen

and liquid oxygen. However, one single process cannot fully utilize LNG cold energy, and other processes that can be combined with air separation should be designed.

In this article, a novel energy integration process for LNG light hydrocarbon separation and air separation has been developed. The optimal temperature ranges of LNG regasification required for the air separation process and the light hydrocarbon separation process are determined via sensitivity analyses.

2. Process design and modeling

2.1 Light hydrocarbon separation unit

Figure 1 shows a schematic diagram of LNG light hydrocarbon separation process. Dapeng LNG receiving terminal in Shenzhen, China is taken as an example (Gu, 2018), where the LNG imports from Australia, that consists of 78 mol% methane, 12.4 mol% ethane, 6.3 mol% propane, 1.4 mol% i-butene, 1.8 mol% n-butane, and 0.1 mol% nitrogen. The LNG is fed at $-162\text{ }^{\circ}\text{C}$ and atmosphere pressure, with a flowrate of 422.4 t/h and firstly pressurized to satisfy the operation pressure of demethanizer. The pressurized LNG is then introduced into the air separation unit to exchange heat with the circulating nitrogen. Next, the LNG stream leaving the air separation unit is sent to the LNG heat exchanger II to condensate the vapor product from the top of demethanizer. After that, the LNG stream flows through a heat exchanger and provides cold energy for the condenser of deethanizer. The heated LNG is finally fed into a demethanizer which works at 1500 kPa. After being partial condensated, the methane-rich gas from the top of demethanizer is sent to a separator. The liquid product from separator is fed to the demethanizer as a reflux, and the gas product is compressed and sent out as pipeline gas. The liquid product from the bottom of demethanizer flows through a throttle valve to reduce its temperature to lower than $-50\text{ }^{\circ}\text{C}$ and fed to a deethanizer. The operating pressure of deethanizer is 110 kPa. The feed of deethanizer is finally separated into ethane and LPG. In this unit, the product lean natural gas contains 99.42 mol% methane, and the product liquefied ethane contains 99.37 mol% ethane.

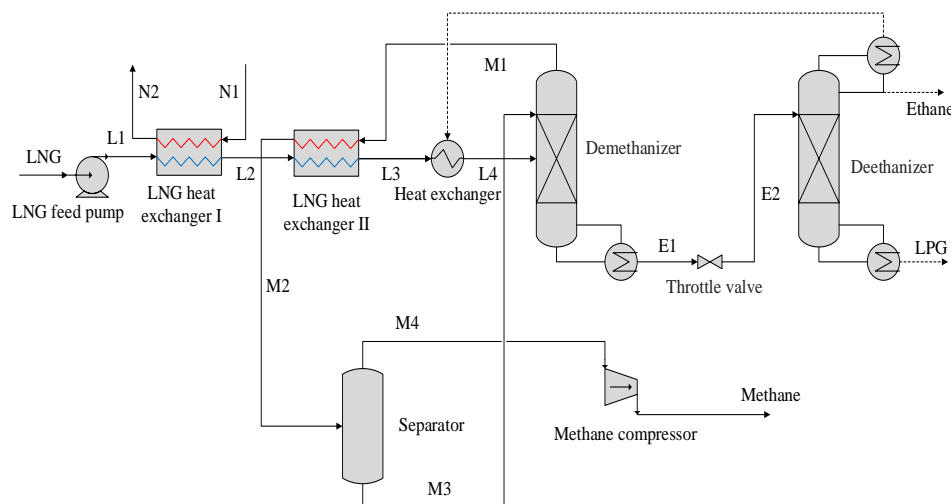


Figure 1: Schematic diagram of LNG light hydrocarbon separation unit

2.2 Air separation unit

Figure 2 illustrates the air separation process, which is adopted from the literature (Xiong and Hua, 2007). The composition of air is simplified to 78.09 mol% nitrogen, 20.95 mol% oxygen and 0.96 mol% argon. The raw air is firstly compressed to 600 kPa by the air compressor and then cooled to $30\text{ }^{\circ}\text{C}$ by the water cooler. Next, the cooled air flows through the LNG heat exchanger II to be further refrigerated. Finally, the air which is at about $-173\text{ }^{\circ}\text{C}$ enters the distillation section. The distillation section of air separation unit mainly contains two distillation columns. The condenser of lower column is energy complementary with the reboiler of upper column, so that the two distillation columns can also be considered as a large distillation column. The air is fed into the lower column from the bottom and the operating pressure of lower column is 550 kPa. The stream N2 is extracted from the middle of lower column as circulating nitrogen. Mixed with stream N3, the circulating nitrogen flows through the LNG heat exchanger II as a coolant, and then enters the cold energy generation section at about $-34\text{ }^{\circ}\text{C}$. The cold energy generation section is a compression refrigeration system using LNG as refrigerant to provide cold utility, including LNG heat exchanger I and two nitrogen compressors. The LNG is heated from -

161.3 °C to -118 °C by the LNG heat exchanger I. The circulating nitrogen is firstly cooled to -130 °C, and then compressed to 1600 kPa by the nitrogen compressors I. After first compression, the circulating nitrogen is cooled to -130 °C again, and further compressed to 5000 kPa by the nitrogen compressors II. Before leaving the cold energy generation section, the circulating nitrogen is finally cooled to -147 °C by the LNG heat exchanger I. Next, the circulating nitrogen flows through the LNG heat exchanger II to be cooled to -172 °C. After that, the circulating nitrogen is depressurized to 560 kPa by a valve and separated into stream N3 and N13. The stream N13 is fed to the lower column as a reflux. After being cooled to about -179 °C by the LNG heat exchanger III, and depressurized to 140 kPa by the valve II, the oxygen-rich gas from the bottom of lower column is sent to the upper column. The nitrogen-rich gas from the top of lower column flows through the LNG heat exchanger III to be cooled to -183 °C. Then, the nitrogen-rich gas is separated into stream N15 and N17. After being depressurized to 120 kPa by the valve IV, the stream N17 is sent to the LN2 tank as liquid nitrogen. There is some boil-off gas from the LN2 tank, namely stream N19. The stream N15 is fed to the upper column after being depressurized to 140 kPa by valve III. The operating pressure of upper column is 120 kPa. The liquid oxygen from the bottom of upper column is sent to the LO2 tank. The waste nitrogen from the top of upper column is mixed with the stream N19, and then flows through the LNG heat exchanger III and LNG heat exchanger II successively to recover residual cold energy. In this unit, the product nitrogen consists of 99.92 mol% nitrogen, 0.03 mol% oxygen and 0.06 mol% argon and the product liquid oxygen consists of 99.72 mol% oxygen and 0.28 mol% argon.

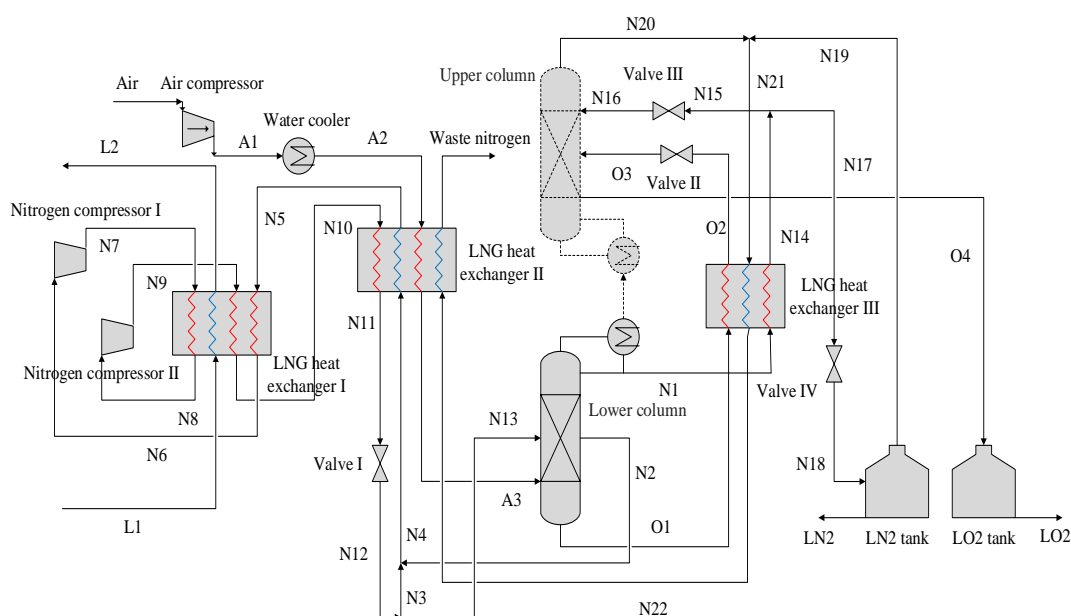


Figure 2: Schematic diagram of air separation unit (Xiong and Hua, 2007)

3. Results and discussion

The process designs for LNG light hydrocarbon separation and air separation are modelled in Aspen Hysys. Three schemes are used for comparison: scheme 1 - only air separation process; scheme 2 - only LNG light hydrocarbon separation process; scheme 3 - energy integrated process of LNG light hydrocarbon separation and air separation. The LNG processing capacity for three schemes is 422.4 t/h.

3.1 Material and energy balance

In this part, the ethane recovery ratio and the utilization ratio of LNG cold energy are the most significant factors. Ethane recovery ratio is an important index to measure the performance of light hydrocarbon separation. The formula for calculating the ethane recovery ratio is given by Equation (1). The results are shown in Table 1.

$$R = \frac{Fx}{F_0x_0} \quad (1)$$

where R is the ethane recovery ratio, F_0 is the LNG flowrate, F is the flowrate of product stream Ethane, x_0 is the ethane composition of LNG and x is the ethane composition of the product stream Ethane.

Figure 1 shows that the temperature of the stream Methane is $-9.703\text{ }^{\circ}\text{C}$. Therefore, the cold energy of LNG is still not fully used. There is no general formula to calculate the utilization ratio of LNG cold energy. The Equation (2) is adopted from the literature (Wang et al., 2015).

$$\eta = \frac{(H_L - H_{L0}) - (H_M - H_{M0})}{H_L - H_{L0}} \quad (2)$$

where η is the utilization ratio of LNG cold energy, H with the subscripts of L , $L0$, M , and $M0$ are the enthalpy of LNG, LNG at reference state, methane and methane at reference state in respective. In the process of heat transfer, the cold energy of LNG depends on the temperature of reference state. The calculation takes the ambient temperature ($20\text{ }^{\circ}\text{C}$) as the reference state, and the results can be found in Table 1.

Table 1: Ethane recovery ratios and utilization ratios of LNG cold energy for different schemes

Scheme	1	2	3
Ethane recovery ratio (%)	-	98.52	97.16
Utilization ratio of LNG cold energy (%)	43.93	79.36	79.70

As shown in Table 1, the ethane recovery ratio of scheme 3 is 97.16%, which is slightly lower than that of scheme 2 (98.52%). But the ethane recovery ratio of scheme 3 also reaches to a significantly high level. The utilization ratio of LNG cold energy of scheme 1 (43.93%) is the lowest. The utilization ratio of scheme 3 is 79.70%, which is slightly higher than that of scheme 2 (79.36%). It indicates that the cascade utilization of cold energy can enhance the usage of LNG cold energy.

3.2 Economic analyses

Economic profit is always a target for process design. It can be expressed as the difference between income and cost. The price of related commodities in China can be found in literatures (Wang et al., 2015) or other references. The income of the process includes lean natural gas (66.90 CNY/t), ethane (6180 CNY/t), LPG (4620 CNY/t), liquid nitrogen (about 650 CNY/t) and liquid oxygen (about 1000 CNY/t). The cost of the process can divide into two sections. Raw material cost is LNG (8.09 CNY/t) and operating cost is power consumption of equipment. The LNG feed pump, the methane compressor, the air compressor, the nitrogen compressors I and the nitrogen compressors II consume electric energy (0.9 CNY/kW·h). Water is used as heat source in reboilers of the demethanizer and the deethanizer, and it costs about 0.23 CNY/kW·h. In addition, the water cooler also needs to consume water, which costs about 0.05 CNY/kW·h. The results of economic analyses are shown in Table 2.

According to the data of economic analyses, the profit of scheme 1 (0.94×10^5 CNY/h) is the least. It is because all the light hydrocarbons that can generate high profits in LNG are wasted. The profit of scheme 3 is 9.20×10^5 CNY/h, which is larger than that of scheme 2 (8.84×10^5 CNY/h). It proves the co-production of light hydrocarbon, liquid nitrogen and liquid oxygen can create larger economic benefits.

3.3 Sensitivity analyses

To optimize this process design, the most important design parameters, temperature of stream L_2 and L_3 in Figure 1 are further investigated through detailed process simulation model in Aspen Hysys. The temperature of stream L_2 is related to the amount of cold energy provided to the air separation unit, and the temperature of stream L_3 determines the amount of cold energy allocated to the condensers of demethanizer and deethanizer. The primary objective of optimization is to maximize the profit, taking into account high ethane recovery ratio and utilization ratio of LNG cold energy.

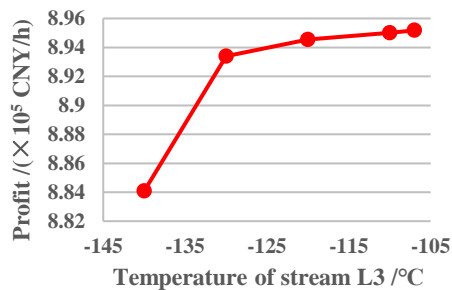
Figure 3 shows profit, ethane recovery ratio and utilization ratio of LNG cold energy over different temperature of stream L_3 . It can be seen that the profit increases as the temperature of stream L_3 changes from $-140\text{ }^{\circ}\text{C}$ to $-107\text{ }^{\circ}\text{C}$. When the temperature of stream L_3 is higher than $-107\text{ }^{\circ}\text{C}$, there is a temperature cross. The ethane recovery ratio increases when temperature of stream L_3 varies from $-140\text{ }^{\circ}\text{C}$ to $-107\text{ }^{\circ}\text{C}$. However, the utilization ratio of LNG cold energy decreases slightly as the temperature of stream L_3 increases. The optimal temperature of stream L_3 is identified as $-107\text{ }^{\circ}\text{C}$ according to the simulation results.

Furthermore, the temperature of stream L_2 is adjusted to optimize the energy integrated process, and the results are shown in Figure 4. The profit increases as the temperature of stream L_2 changes from $-150\text{ }^{\circ}\text{C}$ to $-118\text{ }^{\circ}\text{C}$. However, when the temperature of stream L_2 is above $-118\text{ }^{\circ}\text{C}$, the profit decreases as the temperature of stream L_2 increases. It is because, within a certain range, more cold energy provided to the air separation unit can produce more liquid nitrogen and liquid oxygen. However, if the air separation unit employs too much cold energy, the production of ethane and LPG will decrease due to lack of cold energy supply. In addition, the

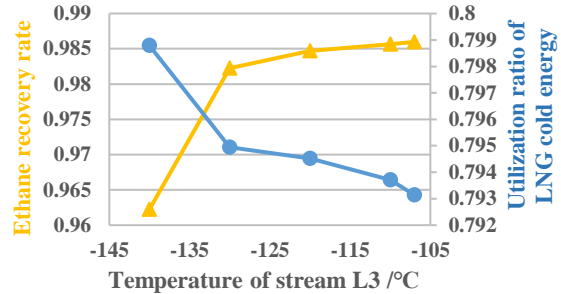
ethane recovery ratio decreases obviously as the stream L₂ changes from -150 °C to -110 °C. The utilization ratio of LNG cold energy increases with the temperature of stream L₂. According to the design specifications, the optimal temperature of stream L₂ is determined as -118 °C.

Table 2: Economic analyses of different schemes

Scheme	1	2	3
Flowrate			
LNG (t/h)	422.4	422.4	422.4
Methane (t/h)	422.4	254.5	255.5
Ethane (t/h)	-	74.78	73.77
LPG (t/h)	-	93.16	93.14
Liquid nitrogen /(t/h)	86.99	-	35.08
Liquid oxygen /(t/h)	83.38	-	33.62
Power			
LNG feed pump (kW)	2181.4	460.87	467.01
Methane compressor (kW)	-	11169	11408
Air compressor (kW)	34291	-	13827
Nitrogen compressors I (kW)	6370.6	-	2568.8
Nitrogen compressors II (kW)	6344.3	-	2558.2
Heat duty			
Reboiler of demethanizer (kW)	-	32731	22811
Reboiler of deethanizer (kW)	-	16574	12255
Water cooler (kW)	32462	-	13090
Economic data			
Income ($\times 10^5$ CNY/h)	1.43	9.10	9.60
Cost ($\times 10^5$ CNY/h)	0.49	0.25	0.40
Profit ($\times 10^5$ CNY/h)	0.94	8.84	9.20

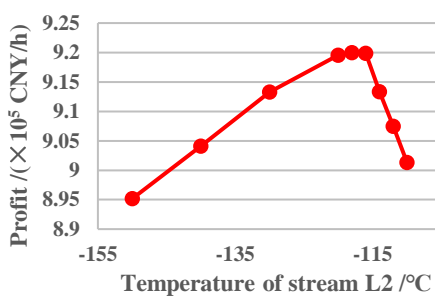


(a)

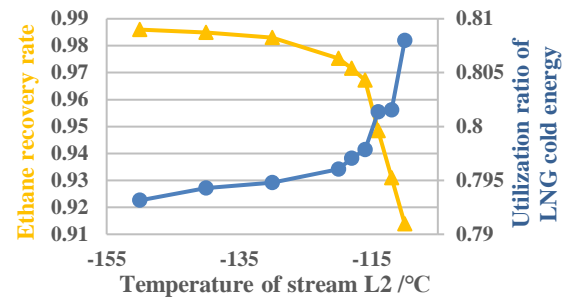


(b)

Figure 3: Influence of the temperature of stream L₃ on (a) profit; (b) ethane recovery rate and utilization ratio of LNG cold energy



(a)



(b)

Figure 4: Influence of the temperature of stream L₂ on (a) profit; (b) ethane recovery rate and utilization ratio of LNG cold energy

4. Conclusions

This work proposed a novel energy integration process for improving the utilization ratio of LNG cold energy and economic profits of the air separation unit in Chinese LNG receiving terminals. To address this technical challenge, the LNG light hydrocarbon separation and cascade utilization of LNG cold energy were adopted. In addition, only air separation process and only LNG light hydrocarbon separation process were designed for comparison. Rigorous simulations for different processes were carried out in Aspen Hysys. Economic analyses and sensitivity analyses were further performed based on the process simulation results. It was found that the profit of energy integration process (9.20×10^5 CNY/h) is the largest among that of only air separation process (0.94×10^5 CNY/h) and only LNG light hydrocarbon separation process (8.84×10^5 CNY/h). Furthermore, the utilization ratio of LNG cold energy of energy integration process is also the highest among the three schemes.

Acknowledgments

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References

- Chen S. Q., Dong X. Z., Xu J., Zhang H. L., Gao Q., Tan C. Q., 2019, Thermodynamic evaluation of the novel distillation column of the air separation unit with integration of liquefied natural gas (LNG) regasification, *Energy*, 171, 341-359.
- Ebrahimi A., Ziabasharhagh M., 2017, Optimal design and integration of a cryogenic Air Separation Unit (ASU) with Liquefied Natural Gas (LNG) as heat sink, thermodynamic and economic analyses, *Energy*, 126, 868-885.
- Fan Q. H., Li H. Y., Yang G. D., Yin Q. S., Wu H., Li S. Y., Jia L. X., 2007, Simulation and analysis of air separation and liquefaction process using LNG cold energy (Chinese), *Sun Yat-sen University Forum*, 27(2), 83-87.
- Gu Z. L., 2018, The simulation and operation optimization of the C2+ recovery process from LNG (Chinese), *Petrochemical Industry Application*, 37(4), 146-149.
- Kim D. H., Giannetta R. E. H., Gundersen T., 2018, Optimal use of liquefied natural gas (LNG) cold energy in air separation units, *Industrial & Engineering Chemistry Research*, 57, 5914-5923.
- Morosuk T., Schult M., Tsatsaronis G., 2014, LNG regasification integrated within an air-separation process, *Proceedings of the ASME 2014 8th International Conference on Energy Sustainability*, 1-11.
- Tesch S., Morosuk T., Tsatsaronis G., 2016, Advanced exergy analysis applied to the process of regasification of LNG (liquefied natural gas) integrated into an air separation process, *Energy*, 117, 550-561.
- Wang Y. F., Li Y. X., Wang W. C., Wang X. S., Liu J. J., 2015, Process of light hydrocarbon recovery from LNG with cryogenic energy utilized in LNG terminals (Chinese), *Chemical Engineering of oil & gas*, 44(3), 44-49.
- Xiong Y. Q., Hua B., 2007, Simulation and analysis of the air separation process by using LNG cold energy (Chinese), *Journal of Shaanxi University of Science & Technology*, 25(3), 68-76.
- Xu W. D., Duan J., Chen Y. W., Zhuo F., 2013, Cold energy utilization technology of LNG and its industrialization progress (Chinese), *Natural Gas Chemical Industry*, 38(5), 79-84.
- Xu W. D., Duan J., Mao W. J., 2014, Process Study and Exergy Analysis of a Novel Air Separation Process Cooled by LNG Cold Energy, *Journal of Thermal Science*, 23(1), 77-84.
- Yamanouchi N., Nagasawa H., 1979, Using LNG cold for air separation, *Chemical Engineering Progress*, 75(7), 78-82.