Optimization of Small-Scale Natural Gas Liquefaction using Nitrogen Expander with Precooled Cycle

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Abstract. Nitrogen expander is the liquefaction process which suitable for small scale liquefied natural gas plant because its characteristic. However, the major issue of technology is low energy efficiency. In this study, optimization of small-scale natural gas liquefaction process using nitrogen expander with precooled cycles were conducted. The specific energy consumption is chosen as an objective function with decision variables are natural gas pressure after compression, nitrogen pressure after compression, inlet temperature of high pressure expander, evaporation temperature in precooling cycle, and type of precooling refrigerant. The refrigerant used for precooling cycle are propane, cyclopropane, isobutane, butane, and neopentane. The simulation and optimization processes are performed by UniSim and GAMS software. The results show that integrating precooling cycle to the nitrogen expander system can reduce specific energy consumption up to 25.24% and propane is the most effective precooling refrigerant.

Keywords: Natural Gas Liquefaction, Small-Scale, Nitrogen Expander, Optimization, Precooling Systems.

1. Introduction
World energy demand is predicted to increase by around 25% from 2016 to 2040 [1]. Due to the abundance and availability of fossil fuel resources, it can be estimated that these resources will continue to play an important role in the world energy economy. Furthermore, fossil fuels currently provide about 85% of the world's commercial energy needs [2].

Natural gas is one of the widely available fossil fuels and produces fewer greenhouse gas emissions than other fuels. It is not surprising that by increasing energy needs and emerging environmental issues, natural gas has become the fastest growing fossil energy source. [3]. Some agencies predict that natural gas consumption in the world will increase by 1.7% per year [4]. Then, as predicted by British Petroleum (BP), the proportion of natural gas will pass through coal in energy consumption by 2035 [5].

In meeting the world's natural gas demand, liquefied natural gas (LNG) is one form of natural gas that supports the logistics of the fuel. The characteristic of LNG which has a volume of about 1/600 of natural gas at room temperature in the same unit of quantity makes it economical to be transported from natural gas sources to the consumers under certain conditions [6]. Furthermore, the process of separating acid gases and other impurities in the LNG plant also produce the high quality of natural gas. Moreover, there is a LNG value chain called small-scale LNG (SSLNG), which one of the roles is meeting natural gas demand in remote areas where there is no conventional natural gas infrastructure. SSLNG is a term of LNG production plant that has capacity of less than 1 million tons of LNG per annum (MTPA) [7]. To date, the total production capacity of SSLNG plant in the world has reached 20 MPTA spread over more than 100 small-scale plant worldwide [8]. The SSLNG will be projected to increase rapidly, reach around 75 to 95 MTPA by 2030 [9]. However, despite of the promising development of this plant, there are several challenges include mainly-cost reductions, engineering design for small scale that safety, supply chain development, and regulations.
The nitrogen expansion liquefaction process is a suitable cycle for SSLNG plant because of its simplicity, fast startup, and easy maintenance [10]. In short, this process utilizes a turbo-expander to produce cooled refrigerants for liquefaction process [11]. However, the cycle gives quite high energy consumption. The mixed refrigerant cycle consumes energy about 46% of nitrogen expander [12].

In the last few years, there are several studies that optimized nitrogen expander liquefaction. He and Ju [10] optimized the nitrogen expander liquefaction process with a precooled cycle system using propane and R410a. As a result, energy consumption of the liquefaction process was reduced by 20.02% and 22.74%, respectively, compared to conventional nitrogen expander liquefaction cycle. Furthermore, Qyumm et al. [13] created an innovation cycle of nitrogen mixed refrigerant with propane and two-phase expander to optimize the nitrogen expander liquefaction process. The energy consumption of the process was reduced by 46.4% compared to nitrogen expander process. Finally, Qyumm et al. [14] proposed an utilization of turbo-expander vortex-tube in the refrigeration system with energy reduction of 68.5% compared to nitrogen expander.

This study aims to optimize the small-scale nitrogen expander liquefaction system with precooling system using hydrocarbon refrigerant group such as propane, cyclopropane, isobutane, butane, and neopentane.

2. Simulation of liquefaction processes
The simulation studies of the proposed LNG process were carried out using a commercial simulator of UniSim Design R390.1. A Peng-Robinson fluid package with the option of Lee–Kesler equation was chosen. It has been investigated that the Lee–Kesler equation is the most accurate enthalpy model for gases especially at higher pressures [15–17]. The design basis dan feed gas conditions are listed in Table 1.

| Properties                          | Condition       |
|-------------------------------------|-----------------|
| Feed condition                      |                 |
| Temperature (°C)                    | 30              |
| Pressure (kPa)                      | 3000            |
| Flow rate (kgmole/hr)               | 6105 (1 Million Ton Per Annum) |
| Natural gas composition             |                 |
| Nitrogen                            | 0.9             |
| Methane                             | 0.04            |
| Ethane                              | 0.02            |
| Propane                             | 0.02            |
| Nitrogen                            | 0.02            |
| Operating conditions                |                 |
| After-coolers outlet temperature (°C) | 40              |
| Liquefaction rate (%)               | 0.95            |
| LNG storage pressure (kPa)          | 200             |
| The adiabatic efficiency of compressor | 0.85           |
| The adiabatic efficiency of expander | 0.8             |

In addition, the following main assumptions were used:
- The suction temperature of precooling fluids compressors should be higher than the dew point temperature
- The minimum internal temperature approach is 2 °C for the LNG heat exchangers.
- The pressure drop across each water-cooler and LNG heat exchangers is negligible
- Heat loss to the environment is negligible
- Water is used as a cooling medium in the interstage coolers of compression units

3. Liquefaction process description
Generally, a conventional nitrogen expander LNG process consists of one loop of refrigerant for the liquefaction of natural gas, including multistage compressions equipped with interstage coolers and
isentropic expansion. The key advantage of N₂ expander based isentropic expansion a process is that they produce the additional energy or useful shaft work that can be integrated with the refrigerant recompression. Figure 1 shows a schematic diagram of the conventional N₂ single expander process.

![Figure 1. Process flow diagram of the conventional nitrogen expander liquefaction process](image1)

Overall, the feed natural gas (stream-11) will be compressed and cooled. Furthermore, the stream will precede the main liquefaction process in cryogenic heat exchanger CHX-1 and CHX-2. Hereafter, the stream will be expanded and enter the separator vessel V-1 so that the pure LNG will be produced (stream-18). Therefore, in the nitrogen liquefaction system, the generated nitrogen will be compressed and cooled, respectively. Furthermore, the nitrogen will be precooled by CHX-1 and expanded twice so the subcooled nitrogen will be produced. Hereafter, the nitrogen will liquefy the natural gas in CHX-2 and CHX-1 respectively and regenerated again hence the cycle is composed. In this study, the temperature of natural gas after first liquefaction process (stream 15) and the pressure ratio is set to 60 °C and 4 respectively.

As explained in previous section, the energy efficiency of this process can be enhanced by introducing a precooling cycle. The process flow diagrams of proposed nitrogen expander liquefaction cycle with precooling system are showed in Figure 2 and 3. The difference between Figure 2 and Figure 3 is the number of compression stage in the precooling system. The reason behind two different models is the refrigerant’s pressure before and after precooling process. For some refrigerants, the values are huge different, so the compression process needs more than one stage in order to keep the pressure ratio below 4. The propane and cyclopropane precooling systems are shown in Figure 2, while the isobutane, butane, and neopentane precooling systems are illustrated in Figure 3.

![Figure 2. Process flow diagram of the proposed nitrogen expander liquefaction process with two compressors in the precooling cycle](image2)
Figure 3. Process flow diagram of the proposed nitrogen expander liquefaction process with one compressor in the precooling cycle

In general, the proposed process flow diagram is quite similar with ordinary nitrogen expander. However, the key difference of the process is an addition of precooling process for the natural gas before entering the main liquefaction process. The precooling system starts with compressing and cooling the generated precooling fluid. The amount of previous process can be done once or twice depends on the precooling fluid’s physical properties. Hereafter, the precooling fluid enters the expansion valve so the fluid becomes cool enough to precool the natural gas in CHX-0. Lastly, the precooling fluid enters the CHX-0 and regenerated again so that a cycle is composed.

4. Optimization Method
For the optimization, the objective is to minimize specific compression energy, with the decision variables of natural gas pressure after compression (stream-12 in Figure 2 and 3), the evaporation temperature in precooling cycle (stream-27 in Figure 2, stream-25 for Figure 3), the nitrogen pressure after compression (stream-4 in Figure 2 and 3) and the inlet temperature of the high-pressure expander (stream-6 in Figure 2 and 3). These decision variables are listed in Table 2. Furthermore, this optimization model can be defined as mixed-integer non-linear programming (MINLP) due to the model has mixed and integer decision variables with subject to the non-linear objective function and linear constraints. The mixed decision variables are \( P_{12} \), \( t_{27/25} \), \( P_4 \), and \( t_6 \) and pre-cooling working fluids act as its integer decision variable. This optimization problem was solved by GAMS and used COUENNE as solver.

Table 2. Decision variables and bounds

| Decision Variables      | Lower Bound | Upper Bound |
|-------------------------|-------------|-------------|
| Stream-12, \( P_{12} \) (kPa) | 4,000       | 8,000       |
| Stream-27/25, \( t_{27/25} \) (°C) | -37         | -30         |
| Propane                 | -28         | -21         |
| Cyclopropane            | -7          | 0           |
| Isobutane               | 4           | 11          |
| nButane                 | 15          | 22          |
| Neopentane              | 2,800       | 3,800       |
| stream-6, \( t_6 \) (°C) | -40         | -35         |

The objective function can be defined as

\[
\text{Min. } f_c(X) = \text{Min.} \left( \sum_{i=1}^{n} \frac{W_{\text{comb}_i} - W_{\text{exp}_i}}{m_{\text{LNG}}} \right)
\]
Let, $f_z(X)$ be specific compression energy of each pre-cooling working fluids $z$ function of vector of decision variables $X$, $W_{com}$ and $W_{exp}$ are power consumed in compressor and power generated in expander respectively, and $m_{LNG}$ infers mass flow rate of LNG.

Response surface methodology (RSM) is selected to represent the model of $f_z(X)$ as the response variables and mixed decision variables as the explanatory variables. The response variable of $f_z(X)$ expressed by $f_z,X$. Hence, the objective function is expressed by Equation 2

$$OF1 = \min \left( \sum_{z \in \mathcal{Z}} a_z f_{z,X} \right)$$  \hspace{1cm} (2)

Here $a_z$ is the integer decision variable. This objective function is constrained to make this optimization feasible with linear constraints. These constraints are:

$$\sum_{z \in \mathcal{Z}} a_z = 1$$  \hspace{1cm} (3)

Where $L$ represents the liquefaction rate of LNG while $\Delta T_1(X)$ and $\Delta T_2(X)$ are minimum internal temperature approach for CHX-1 and CHX-2 respectively.

5. Results and Discussion
5.1 Optimization Result.
The optimization results of nitrogen expanders liquefaction cycle with different working fluid of precooling system are showed in Figure 4.

![Figure 4. Optimization results of small-scale nitrogen expander liquefaction cycle with/without precooling system](image)

Based on Figure 4, it is showed that adding a precooling system to liquefaction cycle I reduces the total energy consumption, where each type of precooling system provides different impact. The nitrogen expander liquefaction cycle with propane-precooled system gives the lowest specific energy consumption of 0.49 kWh/kg comparing to the ordinary nitrogen expander liquefaction cycle of 0.66 kWh/kg. Adding of the precooling system can reduce the specific energy consumption up to 25.24%. Since the precooling systems make the feed natural gas become cooler it reduces the entropy.
generation due to the lower temperature difference between the natural gas and refrigerant in the heat exchangers. Afterwards, the optimum decision variables obtained from optimization are showed in Table 3.

Table 3. Optimum decision variables

| Decision Variables | Optimum Value |
|--------------------|---------------|
| Stream-12, P12 (kPa) | 4,000         |
| Stream-27/25, t27/25 (°C) | Propane -33.90 |
|                      | Cyclopropane -25.10 |
|                      | Isobutane -2.87 |
|                      | Butane 11      |
|                      | Neopentane 11.85 |
| Stream-4, P4 (kPa)  | 2,800         |
| Stream-6, t6 (°C)  | -35           |

Table 3 showed the optimum value of stream-12 and stream-4 are located in the lower bound. Meanwhile, the optimum value of stream-6 is located in upper bound and stream-27/25 is located between lower and upper bound. For the value of stream-12 and stream-4, the key reason behind the result is the energy consumption by compressor becomes lower for the lower value of decision. Therefore, for the stream-6, the energy consumption by compressor and the energy release by expander decreases and increases respectively when the temperature of stream-6 is higher. Lastly, for the stream-27/25, the higher the temperature of stream-27/25 gives, lower pressure difference and lower energy consumption by compressor, but it gives also lower the precooling performance.

5.2 Composite Curve Analysis.
The effect of precooling system with different working fluids can be illustrated in composite curves as shown in Figure 5.
Figure 5 showed that each of liquefaction process has their own typical composite curve. Since the gap between the composite curves represent the entropy generation or exergy loses, the composite curve of liquefaction cycle which has low total energy consumption has the closest gap. Thus, the composite curve of liquefaction cycle with lower total energy consumption such as propane and cyclopropane have tight composite curves and the ordinary liquefaction cycle with high total energy consumption correspond to the wide composite curve.

5.3 Precooling System Analysis.
The total energy consumption for each working fluid of precooling system can be assessed using temperature of natural gas before liquefaction process. The values are showed in Figure 5.

Figure 6 revealed an increasing temperature of natural gas from propane to neopentane. Temperature profile is determined by the cooling capacity of precooling system as well as heat exchanger performance. The phenomena is corresponded with Figure 4, where the hotter temperature of feed natural gives, the higher total energy consumption. The level of temperatures of natural gas determines the mass flow rates and net energy consumption required in nitrogen cycle. Those values are showed in Figure 6.
Based on Figure 7, the mass flow rate and net energy consumption in nitrogen expander cycle with precooling system are increased from propane to neopentane. The increasing mass flow is caused by the increasing of temperature of feed natural gas as shown in the Figure 5. Since the temperature of natural gas is increase, the temperature of nitrogen before evaporation becomes cooler. Hence, the mass flow rate of nitrogen cycle is also increased. As the mass flow rate of nitrogen cycle increase, energy consumption by compressor and energy release by expander in the nitrogen cycle increase. In this case, the different energy release from expander is not high enough to compensate the increase of energy consumption. Thus, the net energy consumption of nitrogen cycle is increased as shown in Figure 7. However, the total energy consumption of liquefaction cycle is not only determined by the net energy consumption of nitrogen cycle, but also by the energy consumption in precooling cycle. The mass flow rate and net energy consumption in precooling cycle are showed in Figure 8.

The mass flow and energy consumption in each precooling system are decreased from propane to neopentane. Since the boiling point of propane to neopentane is increased, the mass flow rate of each precooling system must adjusting by lowering the value. This adjustment must be meet so the precooling system stay in the gas phase. As the mass flow is decreased, the energy consumption of the
A precooling system is also decreased because the compressor needs less energy. Hence, the energy consumption in precooling cycle is decreased. Nevertheless, the reduction of energy consumption in precooling cycle is not high enough to compensate the escalation of net energy consumption in nitrogen cycle.

6. Conclusions
An optimization of small-scale natural gas liquefaction through nitrogen expander was conducted by integrating precooling systems with several common hydrocarbons such as propane, cyclopropane, isobutane, butane, and neopentane as working fluids. The optimization results showed that precooling systems will reduce the total specific energy consumption of liquefaction process up to 25.24% compared to ordinary nitrogen expander liquefaction process. Furthermore, the performance order of five working fluids which ranked from best to worst is propane, cyclopropane, isobutane, butane, and neopentane.

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