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Thermodynamic design of natural gas liquefaction cycles for offshore application

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ABSTRACT

A thermodynamic study is carried out for natural gas liquefaction cycles applicable to offshore floating plants, as partial efforts of an ongoing governmental project in Korea. For offshore liquefaction, the most suitable cycle may be different from the on-land LNG processes under operation, because compactness and simple operation are important as well as thermodynamic efficiency. As a turbine-based cycle, closed Claude cycle is proposed to use NG (natural gas) itself as refrigerant. The optimal condition for NG Claude cycle is determined with a process simulator (Aspen HYSYS), and the results are compared with fully-developed C3-MR (propane pre-cooled mixed refrigerant) JT cycles and various N2 (nitrogen) Brayton cycles in terms of efficiency and compactness. The newly proposed NG Claude cycle could be a good candidate for offshore LNG processes.

1. Introduction

Offshore LNG (liquefied natural gas) production is one of emerging plant markets, including FPSO (floating production storage and offloading). The most suitable thermodynamic cycle for offshore liquefaction of natural gas [1–4] may be different from the on-land processes under operation, such as propane-precooled mixed-refrigerant (C3-MR) process. Thermodynamic efficiency is obviously an important criterion in selecting any liquefaction cycles. According to Barclay and Denton [2], additional factors to consider for offshore liquefaction are compactness, ease of operation, and safety. Furthermore, other constraints in marine environment may be imposed such as vessel motion, modularity of equipment, and small refrigerant inventory.

Turbine-based LNG processes have advantages in these cycle selection criteria over the traditional Joule–Thomson (JT) cycles with MR [1–3]. The simplest cycle to employ a turbine (or an expander) is reverse-Brayton cycle (simply called "Brayton cycle") [1,5,6], as widely used in peak-shaving plants. The refrigerant of Brayton cycle for LNG processes is nitrogen (N2) or mixture of nitrogen and methane [1], since it should remain in gas phase at LNG temperatures. A variety of modifications can be made on standard Brayton cycle to improve thermodynamic efficiency and to more closely meet the requirements of offshore plants, as reported in [7].

Claude cycle is another candidate for offshore LNG process, as commonly used in cryogenic liquefaction plants of nitrogen, oxygen, and hydrogen [8–10]. The historic success of a large-scale helium liquefier by Samuel Collins was accomplished with a serial combination of Claude cycles [8,9]. However, the direct application of open Claude cycle to LNG processes is not so easy, mainly because natural gas (NG) is a mixture of different hydrocarbons and the phase separation at liquid receiver complicates the composition of working fluid. On the other hand, some modifications on Claude cycle may be considered for natural gas liquefaction, especially for offshore application.

This study intends to perform thermodynamic design on various turbine-based cycles and compare the results with the reputed C3-MR process in terms of efficiency and other factors that should be considered for offshore application. An excellent book by Venkatarathnam [1] describes the evolution of LNG processes from simple to very sophisticated processes under operation in large base-load plants. A parallel design basis to the book is constructed in this study for the purpose of selecting offshore LNG processes. This is part of our ongoing efforts supported by the LNG Plant R&D Center under the MOLIT (Ministry of Land, Infrastructure, and Transport) of Korean government.

2. Selection of thermodynamic cycles

2.1. Existing C3-MR JT cycles

Most of LNG processes under operation are based on C3-MR JT cycles, as schematically shown in Fig. 1 [1–3]. The logical starting

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Nomenclature

FOM	figure of merit
h	specific enthalpy (kJ/kg)
$\dot{m}$	mass flow rate (kg/s)
P_H	high-pressure of cycle (MPa)
s	specific entropy (kJ/kg K)
$\dot{S}_{\text{gen}}$	entropy generation rate (kW/K)
T	temperature (K)
$\dot{W}$	power input or output (kW)
x	expander flow ratio

Subscripts

0	ambient
1,2,...	location of cycle
AC	after-cooler
C	compressor
E	expander or turbine
F	feed of natural gas
HX	heat exchanger
LNG	liquefied natural gas
M	mixing
min	minimum
V	JT valve

point of considering a thermodynamic cycle for offshore liquefaction should be this cycle. The NG feed is pre-cooled to around 240 K by four-stage C3 JT cycle, and then condensed and subcooled to LNG temperature by two-stage MR JT cycle. With optimized composition and operating pressure of MR, the combined cycles achieve a great thermodynamic efficiency. It is noted that the expansion in both C3 and MR cycles is basically an "isenthalpic" process through JT valve or simple throttle device.

2.2. Standard and modified N2 Brayton cycles

Brayton cycle is an excellent gas refrigeration cycle including the "isentropic" process with turbine or work-producing expander [1,5–7,9]. The theoretical isentropic process is, however, difficult to closely realize in practice with cryogenic turbines, which is the main reason why Brayton cycle is not so efficient in LNG process. On the other hand, an N2 Brayton cycle has merits for peak-shaving or offshore plants, since N2 is inexpensive, non-flammable, and safe to handle. In practical systems, N2 is compressed to a very high-pressure, typically greater than 10 MPa [1].

Lately, the present authors have presented a thermodynamic study on standard and modified N2 Brayton cycles [7]. The efficiency of standard Brayton cycle shown in Fig. 2(a) can be significantly improved by modifying the cycle in various ways with an additional turbine. Two modified cycles shown in Fig. 2(b) and (c) have been recommended, as called "two-stage" and "dual-turbine" cycles, respectively. In two-stage cycle, two turbines are arranged in series, but in dual-turbine cycle, two turbines are arranged in parallel with the same pressure ratio.

2.3. New proposal – NG Claude cycle

Claude liquefaction cycle is a combined system of isentropic (turbine or expander) and isenthalpic (JT valve) processes. Most of the cryogenic liquefaction plants for nitrogen, oxygen, and hydrogen have been developed upon a basis of open Claude cycle

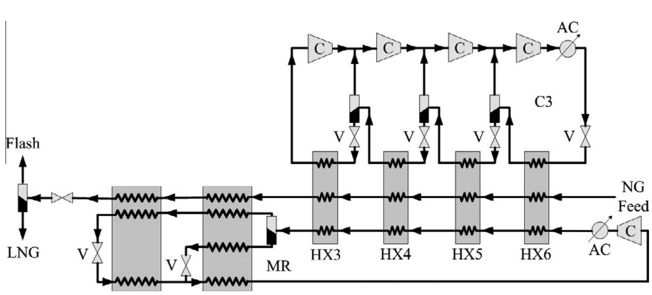


Fig. 1. Propane pre-cooled mixed refrigerant (C3-MR) JT cycle (HX: heat exchanger, *V*: JT valve, *C*: compressor, AC: after-cooler).

[8,9], as shown in Fig. 3(a). The high-pressure gas is branched from the main stream, expanded through an expander, and mixed with the low-pressure return stream. The high-pressure stream to be liquefied is cooled down through heat exchangers and is finally expanded through a JT valve to the liquid receiver.

In order to apply the Claude cycle to natural gas liquefaction, it is proposed to use a closed Claude cycle so that the NG feed is cooled down and liquefied in a separate stream passing through a series of multi-stream heat exchangers, as shown in Fig. 3(b). In addition, it is proposed to use the natural gas (NG) itself as refrigerant in the closed cycle, called "NG Claude Cycle". This cycle may be considered an MR (mixed refrigerant) Claude cycle, whose refrigerant has the same composition as the NG feed. An evident advantage of using NG as refrigerant is to eliminate the need of refrigerant inventory in offshore plants. The first attempt of this study is the thermodynamic design of NG Claude towards its best performance.

3. Simulation and optimization

3.1. Thermodynamic efficiency

The thermodynamic performance of a liquefaction system is evaluated as the work required per unit mass of liquefied gas [1]. By combining the first and second laws of thermodynamics, there exists an absolute minimum in the required work per unit mass, which is expressed as the exergy (flow availability) of LNG [8,9].

$$\dot{W}_{\min}/\dot{m}_F = (h_{\text{LNG}} - h_0) - T_0(s_{\text{LNG}} - s_0) \tag{1}$$

where h and s are specific enthalpy and entropy, respectively, and T_0 is the ambient temperature at which heat is rejected by the liquefaction system.

The thermodynamic efficiency called "figure of merit" (*FOM*) for liquefaction is defined as the minimum input power divided by the actual input power [1].

$$FOM = \frac{\dot{W}_{\min}}{\dot{W}} = \frac{\dot{m}_F[(h_{\text{LNG}} - h_0) - T_0(s_{\text{LNG}} - s_0)]}{\dot{W}_C - \dot{W}_E} \tag{2}$$

In practice, the output power from expander or turbine ($\dot{W}_E$) may be used to help compress the refrigerant or may be dissipated in a brake, but for the purpose of comparison, it is taken into account as negative power in the denominator of Eq. (2) for Brayton and Claude cycles [8].

Thermodynamic irreversibility [9] is the difference between denominator and numerator of Eq. (2), which can be expressed as the sum of entropy generation rate at each component multiplied by the ambient temperature. In the liquefaction systems considered in this study, the components are classified as heat

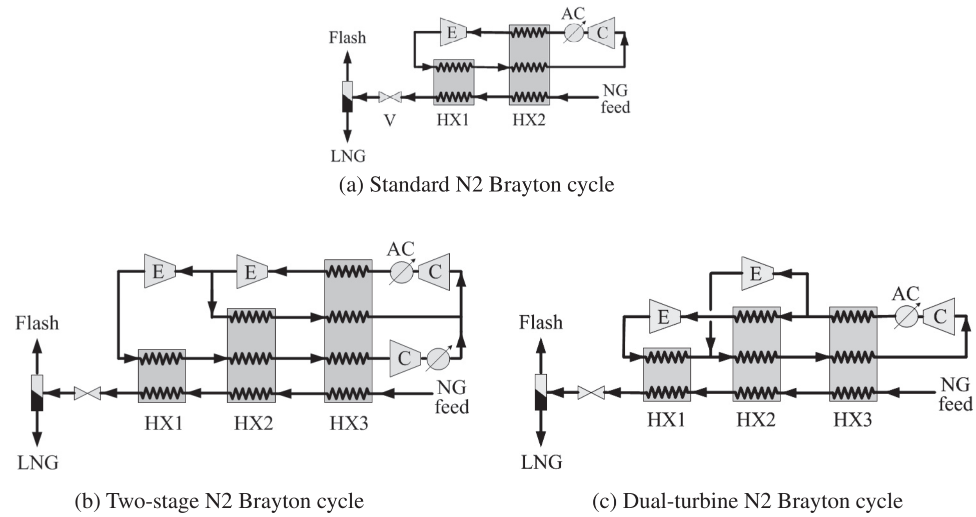


Fig. 2. Standard and modified nitrogen (N2) Brayton cycles (HX: heat exchanger, *V*: valve, *E*: expander, *C*: compressor, AC: after-cooler).

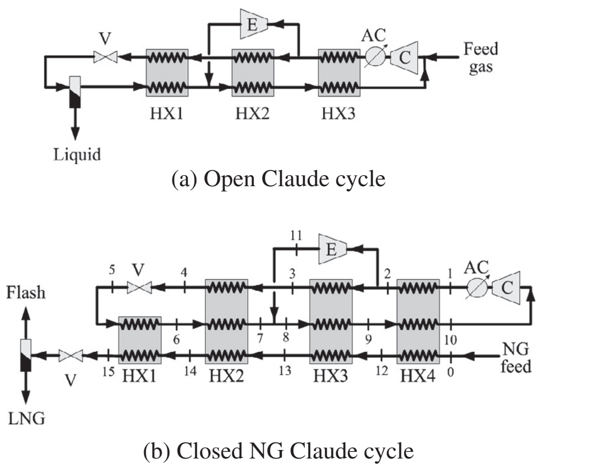


Fig. 3. Open and closed NG (natural gas) Claude cycle (HX: heat exchanger, *V*: JT valve, *E*: expander, *C*: compressor, AC: after-cooler).

exchangers (HX), expanders (*E*), JT valves (*V*), mixing devices (*M*), compressors (*C*), and after-coolers (AC).

$$\dot{W} = \dot{W}_{\min} + T_0(\dot{S}_{\text{gen,HX}} + \dot{S}_{\text{gen},E} + \dot{S}_{\text{gen},V} + \dot{S}_{\text{gen},M} + \dot{S}_{\text{gen},C} + \dot{S}_{\text{gen,AC}}) \quad (3)$$

where the left-handed side is the net input power, and the right-handed side is its expenditure, itemized as exergy increase of NG feed (i.e. the minimum power) and exergy loss contributed by components (the last terms in parenthesis).

3.2. Assumptions and simulation basis

For the present study, the following assumptions are taken in parallel to [1]:

① The composition of natural gas (NG) is 4.0% nitrogen, 87.5% methane, 5.5% ethane, 2.1% propane, 0.5% *n*-butane, 0.3% *i*-butane, and 0.1% *i*-pentane on mole basis.
② The feed pressure of NG is 6.5 MPa.
③ The pressure drop in all heat exchangers is zero.
④ The inlet temperature of NG feed and all refrigerants at the warm-end is 300 K (ambient temperature), and the exit temperature of NG feed at the cold end is 113 K (LNG temperature).
⑤ The maximum pressure of refrigerant is 10 MPa.
⑥ The minimum pressure of refrigerant is 0.13 MPa.
⑦ The adiabatic efficiency of all compressors and expanders is 80%.
⑧ The minimum temperature difference between the hot and cold streams is 3 K in all heat exchangers.

$$\Delta T_{\min} = 3\,\text{K} \quad (4)$$

⑨ The flow rate of NG feed is 1 kg/s for every cycle (for a "unit" liquefaction cycle).

$$\dot{m}_F = 1\ \text{kg/s} \quad (5)$$

A general-purpose process simulator, Aspen HYSYS, is used for the simulation and thermodynamic design of cycles. Thermodynamic properties of NG and refrigerants are calculated with the Peng–Robinson equation of state linked to the simulator. In order to complete the C3-MR JT cycles, a number of design parameters should be determined as well as the assumptions above. The composition and operating pressure of MR are optimized for achieving the highest *FOM* [1]. The pre-cooling temperature and pressure levels of C3 JT cycle are also optimized to minimize the compressor input power. For the purpose of comparison, a reference cycle for C3-MR process is simulated here according to the same method described in [1]. For the standard and modified Brayton cycles, the detailed simulation results presented by the present authors in [7] are also brought for comparison in this paper. It is noted that Assumption ⑥ plays a role in determining the lowest temperature for two-phase refrigerants such as C3 and NG in association with Assumption ⑧. The input power to compressors is calculated with Assumption ⑦ and an additional assumption that the number of compression stages is 4 for C3 and NG, and 3 for MR and N2, respectively [1], so that the pressure ratio at each stage is in the range of 3–4. For multi-stage compression, the intermediate pressures are determined such that the total power input is minimized.

3.3. Optimization of NG Claude cycle

The newly proposed NG Claude cycle is simulated with the assumptions listed above. According to Assumptions ①–④, the states 0 and LNG of NG feed are fixed, and T_1 is also given. In addition, the low-pressure of NG is also fixed at 0.13 MPa with Assumptions ⑥ and ⑧. The number of unknowns is 16, including 2 mass flow rates ($\dot{m}_C$,$\dot{m}_E$), 1 pressure (P_H), and 13 temperatures (T_2 through T_{14}). On the other hand, the number of equations is 14, including 2 for *V* and *M* (energy balance), 1 for *E* (Assumption

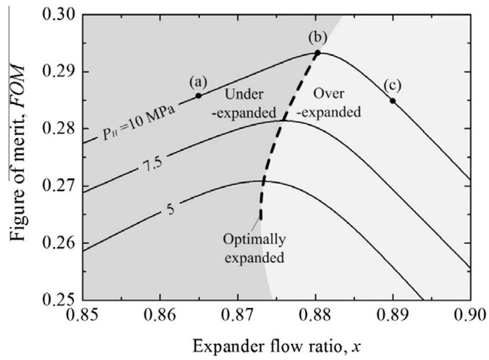


Fig. 4. Simulated *FOM* of NG Claude cycle as a function of x with selected values of P_H, indicating three examples of (a) under-expanded, (b) optimally expanded, and (c) over-expanded condition.

⑦), 2, 3, 3, and 3 for HX1, HX2, HX3, and HX4 (energy balance and Assumption ⑧), respectively. As a result, there remain 2 independent variables to complete the cycle. The first and key parameter in this design is "expander flow ratio" [8], which is defined as the ratio of the mass flow rate passing through expander to the total mass flow rate of compressed refrigerant.

$$x = \frac{\dot{m}_E}{\dot{m}_C} \tag{6}$$

The second parameter is the high-pressure of NG refrigerant (P_H) with a constraint by Assumption ⑤.

Fig. 4 is the plot of simulated *FOM* of NG Claude cycle as a function of x for selected values of P_H. For a given P_H, there exists a unique optimum of x that maximizes the *FOM*. The dashed curve is the locus of the top points of curves, indicated by "optimally expanded" condition. The left and right regions of the curve are

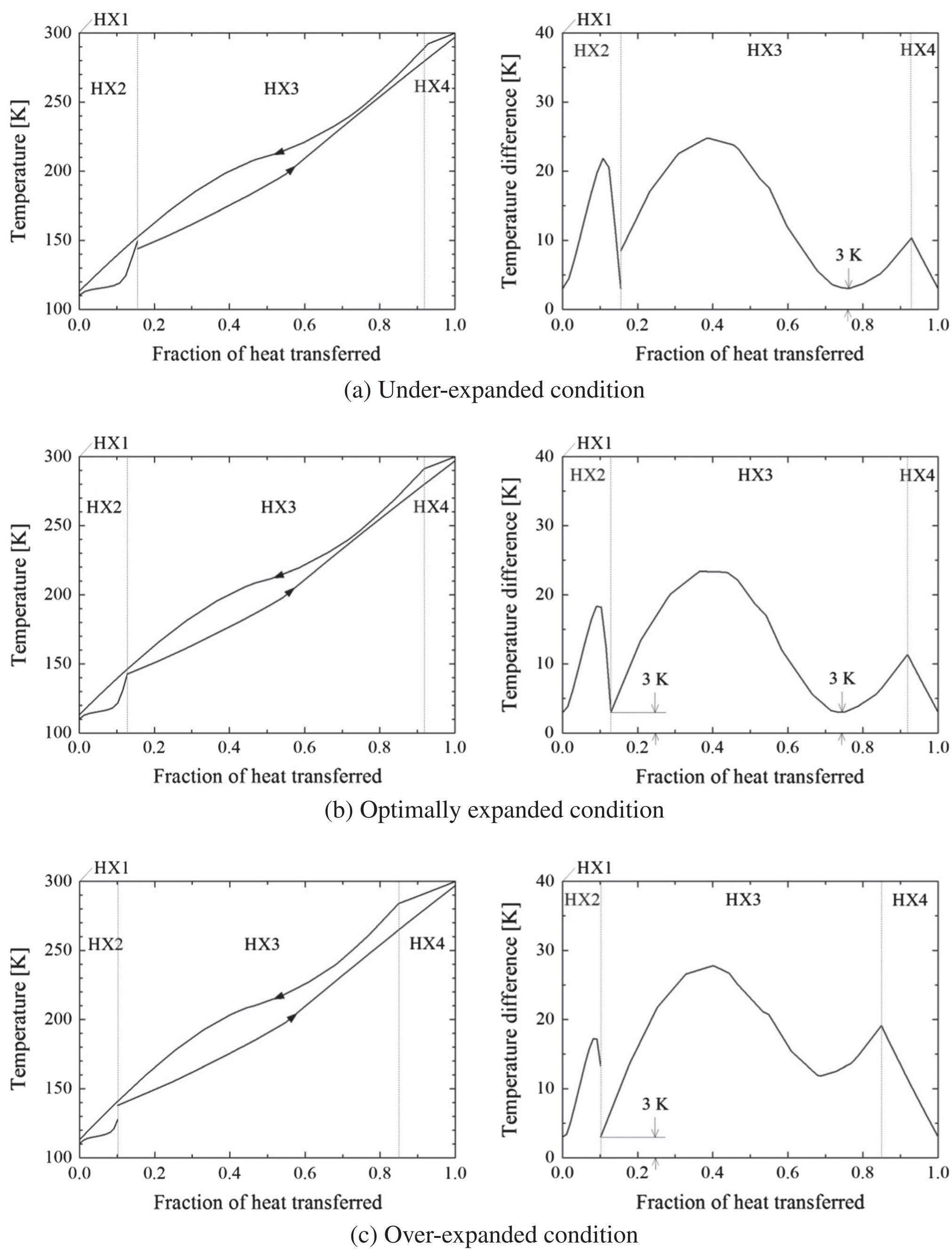


Fig. 5. Temperature profile and temperature difference (ΔT) in HX's.

Table 1
Composition (mole%) of natural gas (NG) for simulation of closed Claude cycle.

		Nitrogen	Methane	Ethane	Propane	*n*-Butane	*i*-Butane	*i*-Pentane
Case 1	Ref. [1]	4.0	87.5	5.5	2.1	0.5	0.3	0.1
Case 2	Qatar	0.4	89.9	6.0	2.2	1.0	0.5	0.0
Case 3	Algeria	0.6	87.6	9.0	2.2	0.4	0.2	0.0

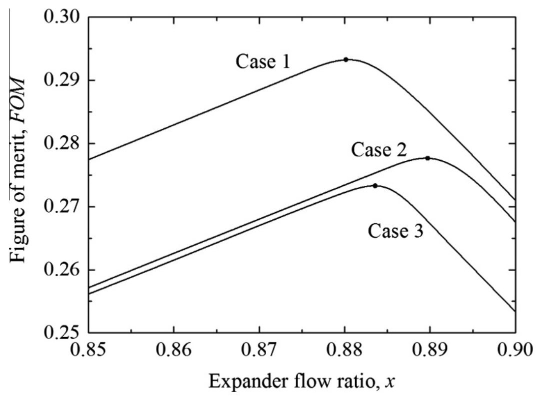


Fig. 6. Simulated *FOM* of NG Claude cycle in different cases of NG composition (P_H = 10 MPa).

called "under-expanded" and "over-expanded" conditions, respectively.

The meaning of optimal expansion can be illustrated by the temperature profiles in HX's, for example, when P_H = 10 MPa. In an under-expanded case (a), the temperature pinch (i.e. the minimum ΔT point) of HX3 occurs near the warm end as shown in Fig. 5(a). On the contrary, in an over-expanded case (c), the temperature pinch of HX3 occurs at the cold end as shown in Fig. 5(c). In the optimally expanded case (b), the temperature pinch occurs at two points so that overall entropy generation due to temperature difference in HX's can be minimized, as shown in Fig. 5(b). This behavior is in a good agreement with the open Claude cycle for liquefaction of pure cryogens [8]. As P_H increases, the maximum *FOM* also increases. In accordance with Assumption ⑤, the cycle is optimized with P_H = 10 MPa and x = 0.880, and the corresponding maximum of *FOM* is 0.294.

The thermodynamic performance of NG Claude cycle may be dependent to an extent upon the composition of NG. The same simulation procedure is repeated for other NG compositions than Assumption ①, as listed in Table 1. Fig. 6 is a plot of simulation results for the three cases with P_H = 10 MPa. The optimal x is the range of 0.88–0.89, and the corresponding maximum of *FOM* varies at 0.27–0.29, depending on the NG composition, especially the N2 concentration. When the NG composition is given in practical offshore plants, it is therefore important to accordingly optimize the expander flow ratio for the best thermodynamic performance.

4. Results and discussion

The simulation results of optimized NG Claude cycle are compared with C3-MR JT cycles and N2 Brayton cycles in terms of efficiency and compactness. First, Fig. 7(a) compares the *FOM*

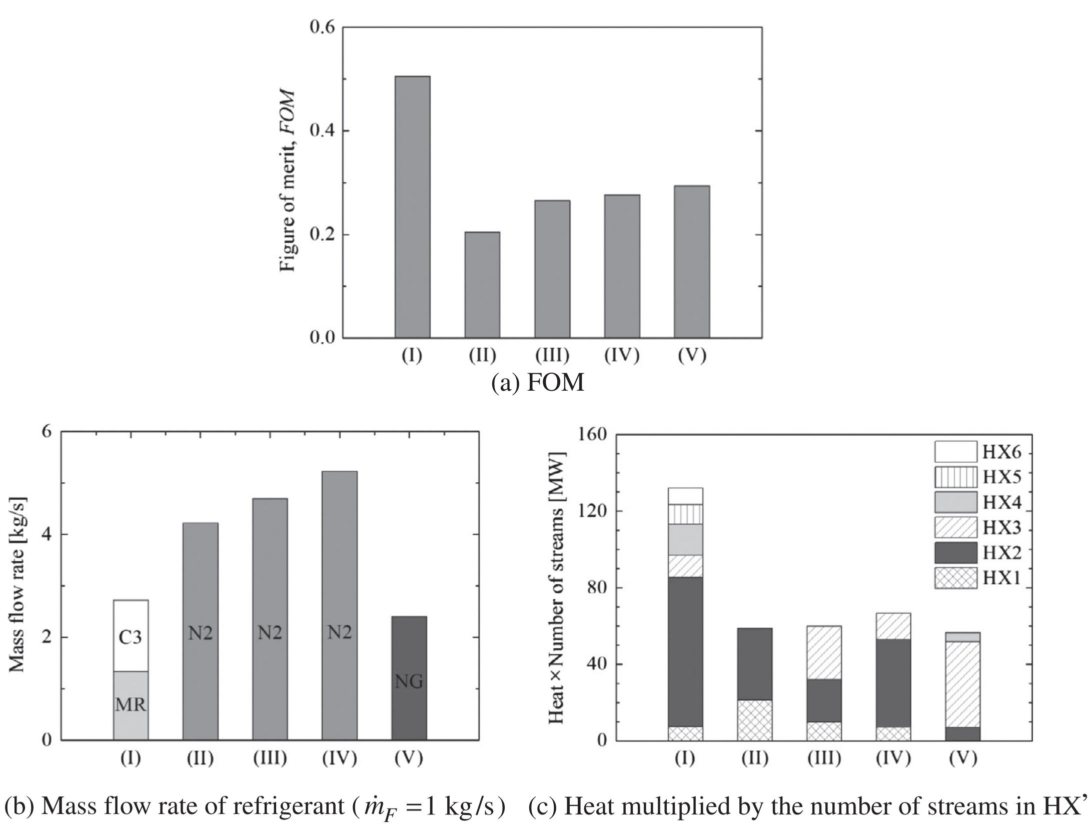


Fig. 7. Comparison of five cycles: (I) C3-MR JT cycle, (II) standard N2 Brayton cycle, (III) two-stage N2 Brayton cycle, (IV) dual-turbine N2 Brayton cycle, and (V) NG Claude cycle.

for the five cycles. The C3-MR JT cycles (I) have a dominantly high *FOM* (50.5%), and the standard N2 Brayton cycle (II) has the lowest *FOM* (20.4%), which are in a good agreement with Sections 6.8 and 6.12 of [1]. Two modified N2 Brayton cycles (III) (IV) and newly proposed NG Claude cycle (V) have a value of *FOM* between 0.265 and 0.294. It is reminded that the modified N2 Brayton cycles (III) and (IV) need two expanders, while the NG Claude cycle (V) requires only one expander like the standard N2 Brayton cycle (II). It is also noted that the *FOM* is calculated with P_H = 10 MPa for Brayton and Claude cycles under the constraint (Assumption ⑤) even though it may reach a slightly higher value if P_H is increased up to 16 MPa.

As mentioned above, the compactness of liquefaction system is another significant factor in selecting a cycle for offshore LNG processes. Fig. 7(b) compares the total mass flow rate of refrigerant through compressor required for the unit mass of LNG ($\dot{m}_F = 1$ kg/s), because it is closely related with the capacity and size of compressors and after-cooling heat exchangers. In case of C3-MR JT cycles (I), the height of column is composed of two mass flow rates of C3 and MR, as indicated by different legends. When compared with C3-MR JT cycles, much more refrigerant flow is required for all N2 Brayton cycles, especially for dual-turbine cycles (IV). The required refrigerant flow rate for NG Claude cycle (V) is well below the one for N2 Brayton cycles (II)–(IV).

Fig. 7(c) compares the sum of heat exchange rates multiplied by the number of streams for all HX's, because this parameter could be an index of estimated size for typical multi-stream heat exchangers used in LNG plants, such as plate-fin or spiral wound HX's. Each column in Fig. 7(c) is stacked up over the serial HX's indicated in Figs. 1–3. The total height of column is shorter in N2

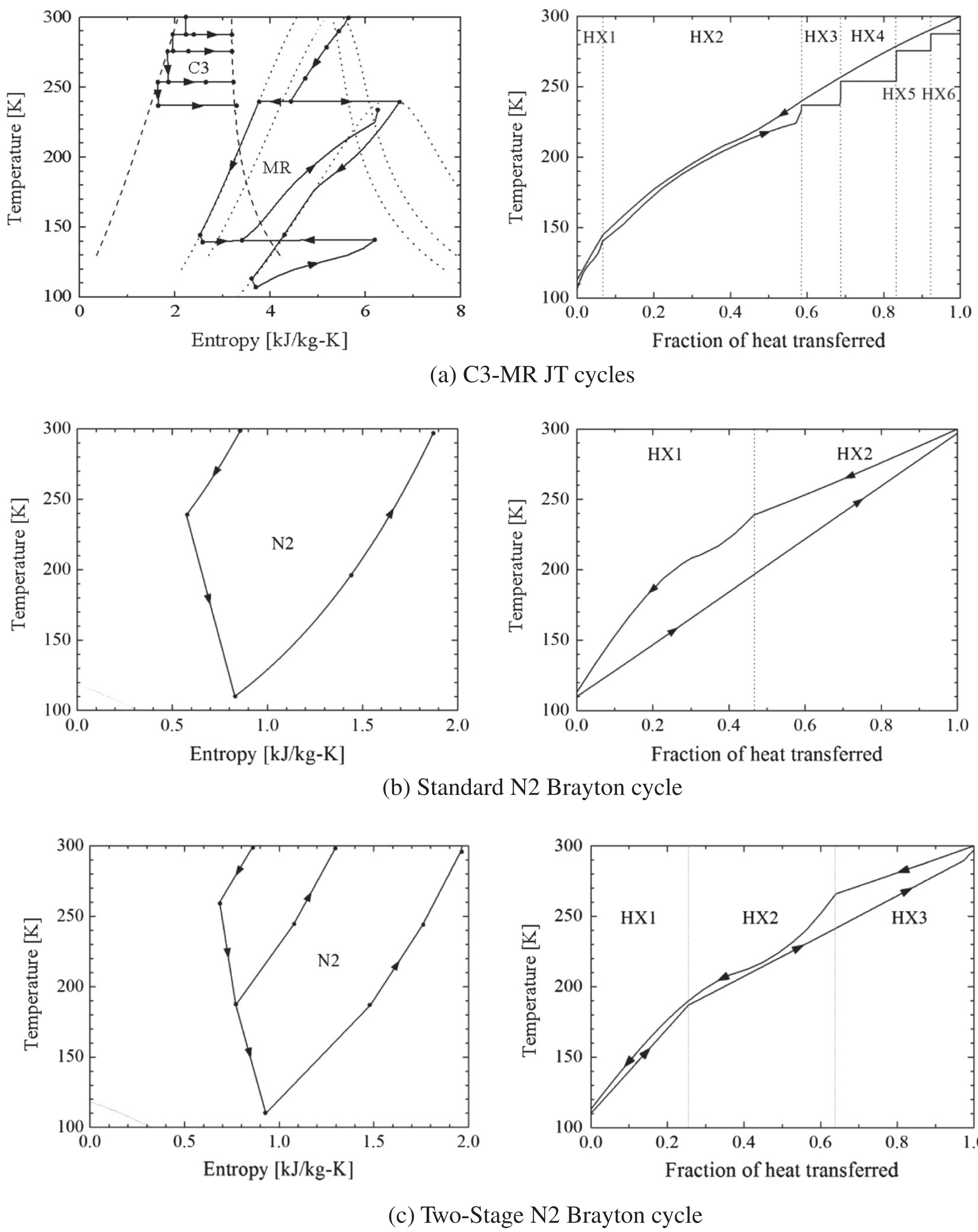
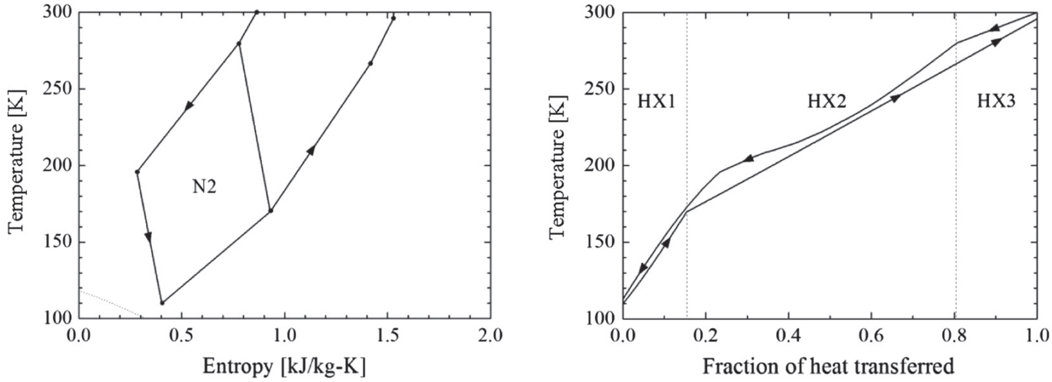
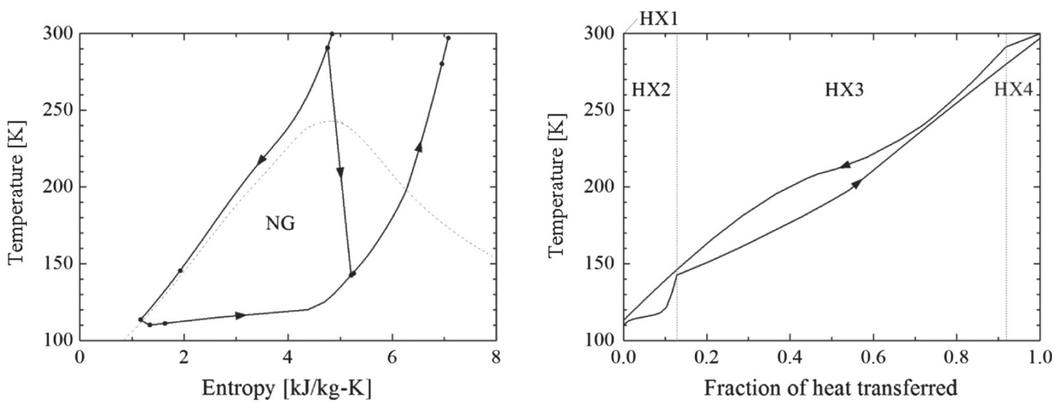


Fig. 8. Temperature-entropy diagram and temperature profile in HX's for five cycles.

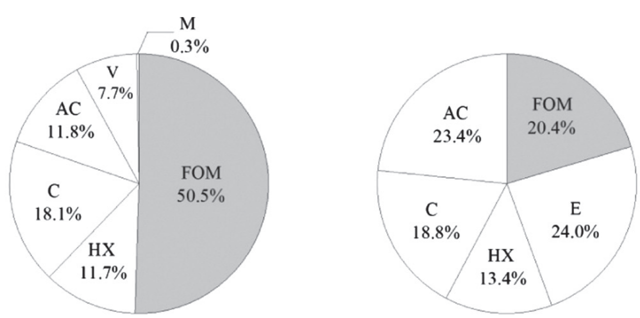


(d) Dual-turbine N2 Brayton cycle

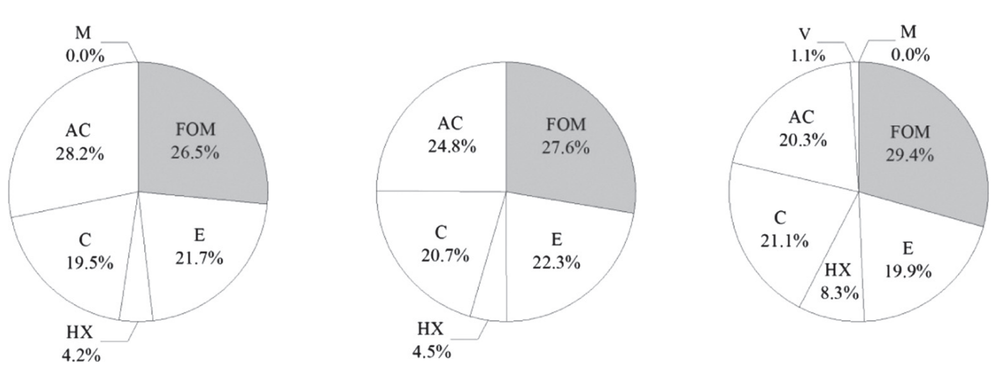


(e) NG Claude cycle

Fig. 8 (*continued*)



(a) C3-MR JT cycles (b) Standard N2 Brayton cycle



(c) Two-stage N2 Brayton cycle (d) Dual-turbine N2 Brayton cycle (e) NG Claude cycle

Fig. 9. Exergy expenditure and irreversibility by parts for five cycles (*FOM*: figure of merit, *E*: expander, HX: heat exchanger, *V*: JT valve, *M*: mixing, *C*: compressor, AC: after-cooler).

Table 2
Temperature, pressure, vapor fraction and mass flow rate of optimized NG Claude cycle.

	Temperature (K)	Pressure (MPa)	Vapor fraction	Flow rate (kg/s)
NG refrigerant				
1	300.0	10.0	1.000	2.408
2	291.4	10.0	1.000	2.408
3	146.4	10.0	0.000	0.289
4	113.3	10.0	0.000	0.289
5	110.0	0.13	0.061	0.289
6	110.3	0.13	0.068	0.289
7	143.4	0.13	0.925	0.289
8	142.7	0.13	0.924	2.408
9	280.1	0.13	1.000	2.408
10	297.0	0.13	1.000	2.408
11	142.6	0.13	0.923	2.119
NG feed				
0	300.0	6.50	1.000	1.000
12	291.4	6.50	1.000	1.000
13	146.4	6.50	0.000	1.000
14	113.3	6.50	0.000	1.000
15	113.0	6.50	0.000	1.000

Brayton and NG Claude cycles (II)–(V), when compared with C3-MR JT cycles (I). Among four candidate cycles (II)–(V) for offshore plants, the standard N2 Brayton cycle (II) and the NG Claude cycle (V) are expected to be the most compact in HX size.

Overall, the newly proposed NG Claude cycle (V) is superior to N2 Brayton cycles (II)–(IV) from the point of both efficiency and compactness, and therefore could be quite suitable for offshore LNG processes. It is recalled that the NG Claude cycle does not need any refrigerant inventory. On the other hand, the NG Claude cycle has an inherent weakness in safety and simplicity over the N2 Brayton cycles, because the flammable refrigerant should be compressed to a high pressure and the expander flow ratio should be carefully optimized in order to achieve the best thermodynamic performance.

For comparing the thermodynamic characteristics of the five cycles, temperature–entropy (T–s) diagram and temperature profile in HX's are presented in Fig. 8. On T–s diagram of MR in Fig. 8(a), there are three saturation (dotted) curves, since the MR has different compositions before and after the phase separator. In Fig. 8(b), the temperature difference is large in the middle, as a single-component (N2) refrigerant is used for a simple cycle. In Fig. 8(c)–(e), however, the temperature difference in HX's is made smaller with different flow rates for hot and cold streams by branching and mixing the streams.

Another crucial issue in thermodynamic design of Claude cycle is the possibility of partial condensation of refrigerant at the exit of expander, as the liquid drops can cause erosion to the high-speed rotor. It is reported that the mass fraction of vapor should be 0.85 or larger to minimize the damage [10]. The vapor fraction at the exit of expander is 0.923 for the optimized cycle, which meets this design constraint. The practical design of expander is beyond the scope of this thermodynamic design, but it can be mentioned that the expansion process may be composed of multiple turbines in parallel and/or in series. For example, a heavy mass flow is branched into multiple circuits in parallel with the same turbines, and a high pressure ratio is multi-staged in series. In any case, the thermodynamic cycle can be reasonably modeled with a value of adiabatic efficiency for expansion.

The thermodynamic irreversibility in the liquefaction systems is the difference between net exergy input and minimum input power, which can be itemized by components in Eq. (3). Fig. 9 shows the exergy expenditure and irreversibility by parts for the five cycles. The total irreversibility fraction is almost identical for three cycles (c)–(e), but the contribution by components is different each other. The HX irreversibility is smaller in (c) and (d), as noticed by the smaller temperature difference in Fig. 8. On the other hand, the E irreversibility is smaller in (e), because the NG Brayton cycle has only one expander. Since the expander flow ratio is optimized, the irreversibility by mixing is negligibly small for all three cycles.

In summary, the details of optimized NG Claude cycle are presented in Table 2, including the temperature, pressure, vapor fraction (in mass), and flow rate of the refrigerant and feed at each location of cycle. It may be confirmed that all the assumptions and design constrains are satisfied. It is noted that the role of HX1 in the optimized NG Claude cycle seems insignificant, but the coldest HX is needed for completing and comparing cycles upon the same simulation basis (especially, Assumptions ⑥ and ⑧). In practice, HX1 and HX2 can be fabricated simply in a piece of heat exchanger (such as plate-fin HX or spiral-wound HX), where the exit tube of high-pressure NG stream (state 4) is located at a slightly warmer location above the cold end.

5. Conclusions

Turbine-based natural gas liquefaction cycles are thermodynamically investigated for application to offshore floating plants. Four candidate cycles are selected and simulated with commercial software, Aspen HYSYS, and the results are compared quantitatively with the fully-developed C3-MR (propane-precooled mixed refrigerant) JT cycles in terms of thermodynamic efficiency and compactness. In particular, a closed Claude cycle is newly proposed to use the NG (natural gas) itself as refrigerant. It is revealed that the NG Claude cycle with one turbine could be as efficient as modified N2 (nitrogen) Brayton cycles with two turbines, and could be more compact in the overall size of liquefaction system. Details of optimized NG Claude cycle are presented for further development.

Acknowledgments

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