Modified Reverse-Brayton Cycles for Efficient Liquefaction of Natural Gas

H.M. Chang[1], J.H. Park[1], K.S. Cha[2], S. Lee[2] and K.H. Choe[2]

[1]Hong Ik University, Seoul, Korea 121-791
[2]Korea Gas Corporation, Incheon, Korea 406-130

ABSTRACT

A thermodynamic study on a variety of modified reverse-Brayton cycles is carried out, aiming at efficient and compact liquefaction systems for natural gas. Reverse-Brayton cycles with nitrogen turbines have been used as peak-shaving LNG processes on a small scale and are also under consideration for application to the lately emerging market for off-shore plants on a larger scale. The standard cycle with one turbine can be modified by employing two turbines in different ways and by adding a pre-cooling Joule-Thomson (JT) stage. The standard and modified cycles are simulated with a commercial process simulator (Aspen HYSYS) based upon selected specifications. Twenty five different cycles are compared in terms of figure of merit (FOM) and the estimated size of heat exchangers. Two-stage expansion cycle and dual-turbine cycle are recommended for efficiency and compactness.

INTRODUCTION

Over decades, a number of thermodynamic cycles[1-3] have been investigated and developed for the liquefaction process of natural gas in order to meet the demand for high efficiency and simple equipment. Since natural gas is a mixture of different hydrocarbons, the specific heat varies considerably with temperature along the liquefaction process. For efficient liquefaction, it is important to reduce the entropy generation due to the temperature difference in heat exchangers[4]. In general, mixed refrigerant (MR)[3] is effective in reducing the temperature difference with a small number of component items in Joule-Thomson (JT) cycles. The reverse-Brayton cycles[3-4] are turbine-based processes that have been used for peak shaving on a small scale, taking advantage of simplicity and quick start-up. Recently, the reverse-Brayton cycles are also under consideration for application to the lately emerging market for off-shore plants on a larger scale, including the on-board liquefaction on LNG ships or FPSO (floating production, storage and offloading) systems.

The reverse-Brayton cycle is an excellent gas refrigeration cycle that is composed of isentropic processes (compression and expansion) and isobaric processes (heat exchangers). The isentropic process is, however, difficult to realize in practice with contemporary cryogenic turbines, which is the main reason why its thermodynamic efficiency is considerably lower than typical JT cycles with MR. On the other hand, reverse-Brayton cycles have some advantageous features for peak-shaving or off-shore applications, as the common refrigerant is nitrogen gas.

Nitrogen is inexpensive, non-flammable, and safe to handle even at a high pressure, typically greater than 100 bar. In addition, the nitrogen cycle is a single-component system that can be easy, simple, and robust in operation.

This study intends to identify and evaluate thermodynamically a variety of natural gas liquefaction cycles with one or two turbines. A basic reverse-Brayton cycle with one turbine is considered first, and then different combinations of two turbines are devised to complete an efficient thermodynamic cycle. It is also quantitatively examined how much the liquefaction performance can be improve by adding a pre-cooling JT cycle at the warm end. A variety of cycles are simulated with commercial software based upon selected design specifications, and are compared in terms of liquefaction efficiency and the estimated heat exchangers size.

STANDARD AND MODIFIED CYCLES

The standard and modified reverse-Brayton cycles under investigation are shown in Fig. 1. Fig. 1(I) is the standard cycle with one turbine and two heat exchangers in series (HX1 and HX2), where the natural gas (NG) feed is cooled from ambient temperature (300 K) to the LNG temperature (113 K). As two turbines are employed, four different modifications are considered in this study. Fig. 1(II) is called "two-stage expansion" cycle, where two turbines are arranged in series and the nitrogen stream is branched to the regenerative heat exchanger after the first expansion. Fig. 1(III) and 1(IV) are called "dual-turbine" cycles, where two turbines are arranged in parallel and the compressed nitrogen gas is branched into two turbines before expansion. The difference between Fig. 1(III) and 1(IV) is whether the two turbines have the same pressure ratio (III) or not (IV). Fig. 1(V) is called a "dual-cycle", where two separate cycles are arranged in parallel for cooling and liquefying an NG feed stream.

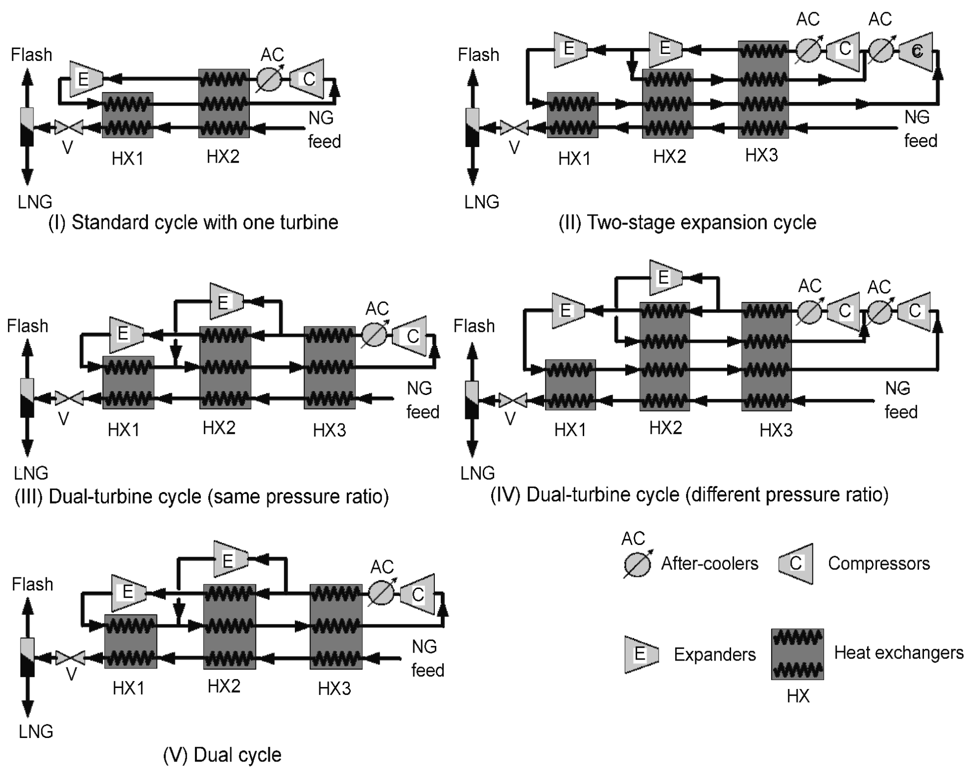


Figure 1. Reverse-Brayton cycles: (I) Standard cycle with one turbine, (II -V) Modified cycles.

The thermodynamic performance of liquefaction systems shown in Fig. 1 may be improved with the assistance of a pre-cooling cycle. There are a number of different options in the pre-cooling cycle, as shown in Fig. 2. In typical LNG processes, the pre-cooling cycle plays a role to cool the NG feed in vapor phase from 300 K to around 240 K, which is close to its saturation temperature. The most popular pre-cooling option under operation is to use multi-stage JT cycles with two-phase refrigerant such as propane (C3) or ammonia (NH_3). Since these are single-component refrigerants, the pre-cooling cycle has typically three (or four) evaporating levels as shown in Fig. 2(a). The number of pre-cooling stages may be reduced, if mixed refrigerant (MR) is used in a JT cycle as shown in Fig. 2(b). It is important, however, that the composition and pressure should be carefully optimized to achieve the improved efficiency with MR cycles[3]. A new concept of pre-cooling cycle has been patented recently by the authors to pursue the efficiency and simplicity at the same time. A parallel combination of ethane and butane (C_2+C_4) JT cycles[5] shown in Fig. 2(c) could be as efficient as the optimized MR cycle, while still taking advantage of easy operation with single-component refrigerants.

There are two ways to locate the low-pressure N_2 stream in the pre-cooled systems. First, the low-pressure (or cold) N_2 stream passes through the pre-cooling HX such that the N_2 cycle is fully regenerative. Second, the low-pressure N_2 stream is directly fed into cold compressors as shown in Fig. 2. The two cycles are comprehensively investigated in the authors' previous work[6], revealing that even though the former might be slightly more efficient, the latter is preferred because of compactness and simplicity. In this study, therefore, only the second way is presented in this paper, as shown in Fig. 2. Similar pre-cooling systems corresponding to modified cycles in Fig. 1(II) through (V) are also examined here, but the schematic diagrams of cycles are not illustrated due to space limitation. In summary, five different cycles (I)-(V) and their pre-cooled cycles with four different options are under consideration in this study. Thus, a total of 25 cycles are simulated and compared.

CYCLE SIMULATION

In order to make a fair comparison of the cycles, the following assumptions are made in similar ways presented by Venkatarathnam[3]:

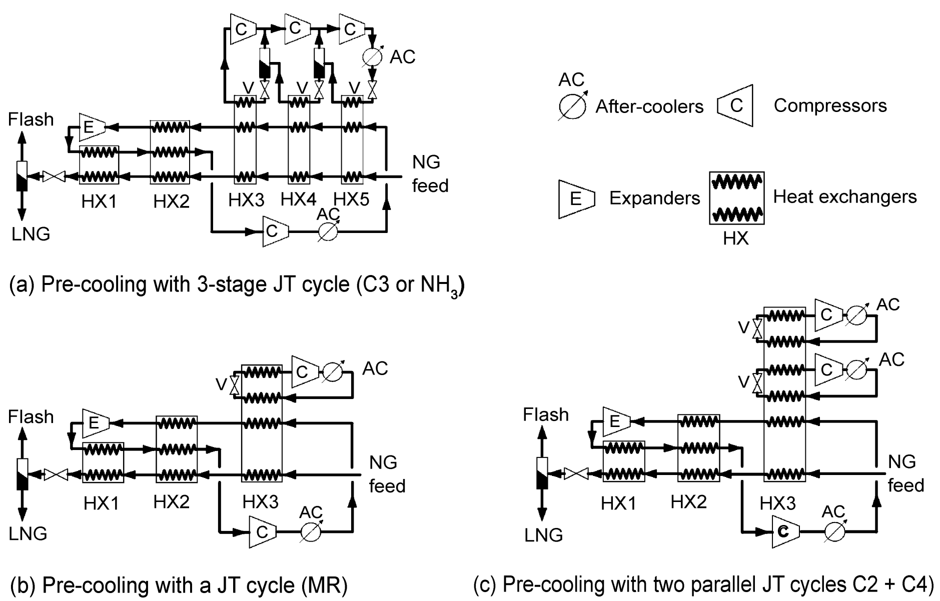


Figure 2. Pre-cooling options of standard reverse-Brayton cycle (I).

- The pressure drop in all heat exchangers is zero.
- The composition of natural gas (NG) feed 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.
- The NG feed is cooled from 300 K (ambient temperature) to 113 K (LNG temperature) at 65 bar for flash expansion to LNG storage.
- The exit temperature of after-coolers (or the inlet temperature of cold box) is 300 K for N_2 cycles, 315 K for C_3 and NH_3 cycles, and 310 K for (C_2+C_4) and MR cycles.
- The minimum temperature approach between hot and cold streams is 3 K in all heat exchangers.
- The adiabatic efficiency of all turbines is 80%.
- The minimum pressure of all refrigerants is 1.0 bar
- The maximum pressure of nitrogen gas is 100 bar.
- The pre-cooled temperature of NG feed is 240 K for all pre-cooling cycles.

Aspen HYSYS Version 7.3 is used for simulation. The composition of pre-cooling MR and the flow rates and pressure levels of combined pure refrigerants (C_2+C_4) are optimized[3,5] subject to the given constraints.

The actual compression work is difficult to estimate, mainly because it depends upon the number of compression stages as well as their efficiency. In order to focus on the thermodynamic nature of various liquefaction cycles, the figure of merit (*FOM*) of a liquefaction system is defined as the ratio of minimum work (the exergy or flow availability of LNG) to the exergy expenditure (the exergy input to the cold box subtracted by the expansion power output)[3,7]

$$FOM = \frac{\dot{m}_F\left[(h_L - h_0) - T_0(s_L - s_0)\right]}{\sum_R \dot{m}_R\left[(h_i - h_e) - T_0(s_i - s_e)\right] - \sum_E \dot{W}_E} \tag{1}$$

where *h* and *s* are specific enthalpy and entropy, respectively. The subscripts *L* and *0* denote the LNG temperature (113 K) and ambient temperature (300 K) of feed gas (*F*), respectively, and the subscripts *i* and *e* denote the inlet and exit of cold box for refrigerants (*R*), respectively. In the denominator of Eq. (1), the exergy input should be the sum for nitrogen gas (N_2) and (if any) pre-cooling refrigerants, and the expansion power output should be the sum for all turbines (*E*).

RESULT AND DISCUSSION

In order to confirm the validity, the simulation result for the standard cycle (I) is compared with the data presented by Venkatarathnam[3]. Fig. 3 is the liquefaction process on the temperature-entropy (T-s) diagrams and the temperature profile in two heat exchangers, which are in good agreement with the results in Section 6.12[3]. The FOM is calculated to be 35.4% from Eq. (1), which is slightly higher because of the different pressure condition.

Fig. 4 is a bar graph to compare the calculated FOM for all 25 cycles. The main refrigerant is nitrogen (N2) for all standard and modified cycles, and the pre-cooling refrigerant is C_3, NH_3,

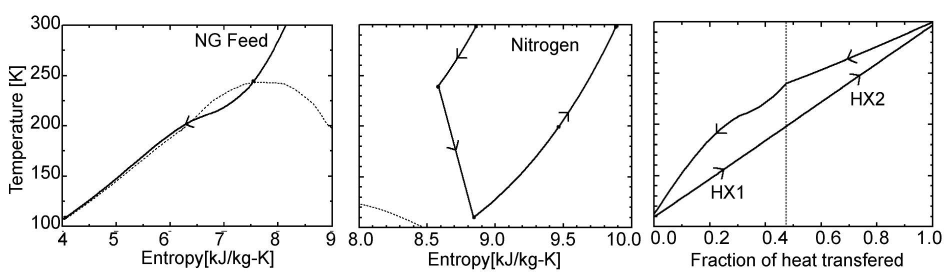


Figure 3. Standard reverse-Brayton cycle (I) on temperature-entropy diagrams (NG feed and nitrogen) and the temperature profile of hot and cold streams in heat exchangers.

MR and (C_2+C_4), respectively. The composition of pre-cooling MR is 44.7% (C_2), 13.0% (C_3), 42.3% (C_4) for pre-cooling (I) and 41.5% (C_2), 18.9% (C_3), 39.6% (C_4) for pre-cooling the cycles (II) through (V). The pressure level and flow rate of pre-cooling refrigerants are optimized as described in Chang et al.[5,6] There is a significant increases in FOM (as much as 15%), if the standard cycle (I) is modified with two turbines in any ways of (II) through (V). It is also noticed that the thermodynamic performance of each cycle may be improved by adding a pre-cooling cycle, but the increase in FOM does not exceed 6%. It can be mentioned that any type of two-turbine cycles with no pre-cooling are superior to any pre-cooled one-turbine cycle. Among the pre-cooling options, the MR cycle yields the most margins for all five cycles (I)-(V), as far as the FOM is concerned.

The size or weight of liquefaction system is another factor in designing the liquefaction process, especially in off-shore or on-board plants. Fig. 5 compares the sum of heat exchange rates multiplied by the number of streams for 25 cycles. This parameter is considered an index to estimate the total size of heat exchangers, even though the actual size may be dependent on detailed (plate-fin or spiral-wound) specifications. Among the group (I), the total stocked column

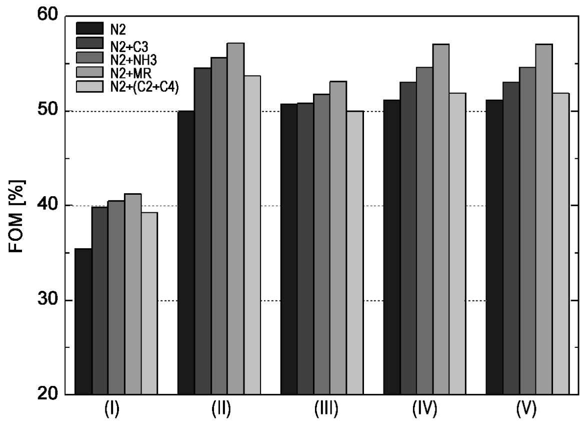


Figure 4. Calculated figure of merit (FOM) for various reverse-Brayton cycles.

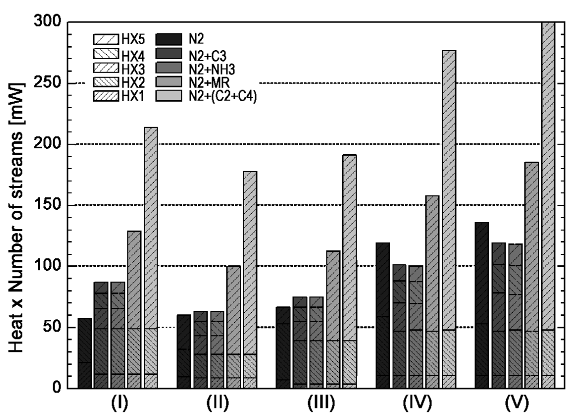


Figure 5. Heat exchange rate multiplied by the number of streams to compare the estimated size of heat exchanger for various reverse-Brayton cycles.

is shortest in basic cycle without pre-cooling and taller in pre-cooled cycles. This means that if a pre-cooling cycle is added, the size of basic cycle should become greater in size as a penalty of the increased FOM. For the two-turbine cycles (II)-(V), on the other hand, the height of columns for pre-cooled cycles with C_3 and NH_3 are not much taller (or even shorter) than the cycles with no pre-cooling. In other words, when a pre-cooling cycle is added, the required size of main (N_2) heat exchanger is reduced for all cycles with two turbines. It is noted that both C_3 and NH_3 are excellent pre-cooling refrigerants, but NH_3 may be selected for off-shore platforms where a small hydrocarbon inventory is preferred from the safety point of view. It is noted that the penalty of large size is severe for the pre-cooling with MR and (C_2+C_4). In summary, the type of main N_2 cycles and pre-cooling cycles should be carefully determined, taking into account the thermodynamic efficiency (Fig. 4) and expected compactness (Fig. 5) at the same time.

For better understanding the thermodynamic nature of simulated cycles, the irreversibility by components is presented. Combining the energy and entropy balance equations[7], the exergy balance of liquefaction cycle can be written as

$$\sum_R \dot{m}_R\left[\left(h_i-h_e\right)-T_0\left(s_i-s_e\right)\right]-\sum_E \dot{W}_E=\dot{m}_F\left[\left(h_L-h_0\right)-T_0\left(s_L-s_0\right)\right]+T_0\sum \dot{S}_{gen} \qquad (2)$$

where the left-handed side is the net exergy input for liquefaction, and the bracket in the right-handed side is the useful effect (i.e. the exergy increase of feed gas). The ratio of useful effect to power input is the FOM as defined in Eq. (1). The last term is the difference between the exergy input and useful effect, which is the irreversibility or the total entropy generation rate multiplied by ambient temperature. The contribution of irreversibility can be itemized for all components in five cycles (I)-(V) and illustrated in Fig. 6. In addition, the four cycles (II)-(V) are illustrated and compared in Fig. 7 as temperature-entropy diagrams and temperature profile in the heat exchangers. It is recalled that the cycles only in the cold box are displayed and that the minimum temperature approach is 3 K for all heat exchangers.

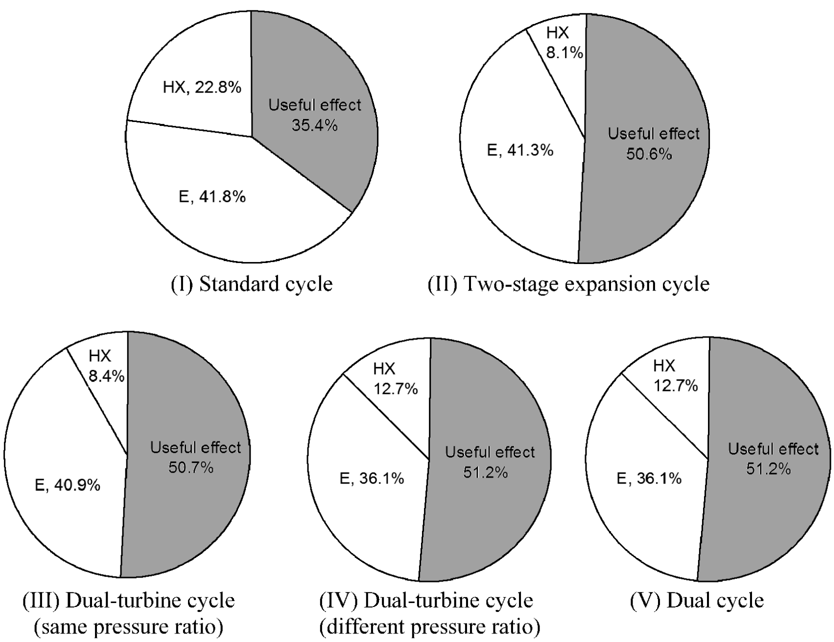
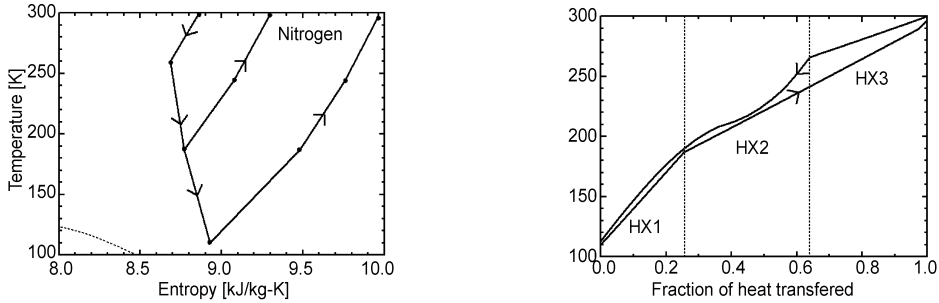
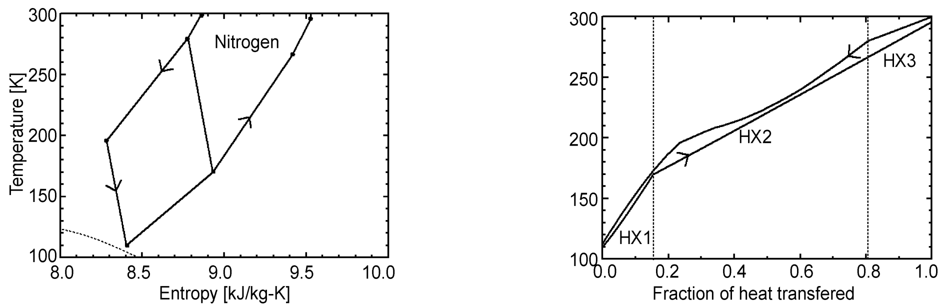


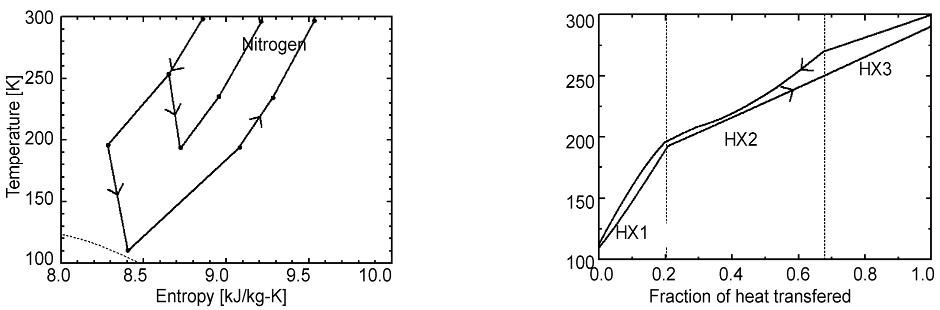
Figure 6. Irreversibility by components in cold box for standard and modified cycles. (HX: heat exchangers, E: expenders or turbines)



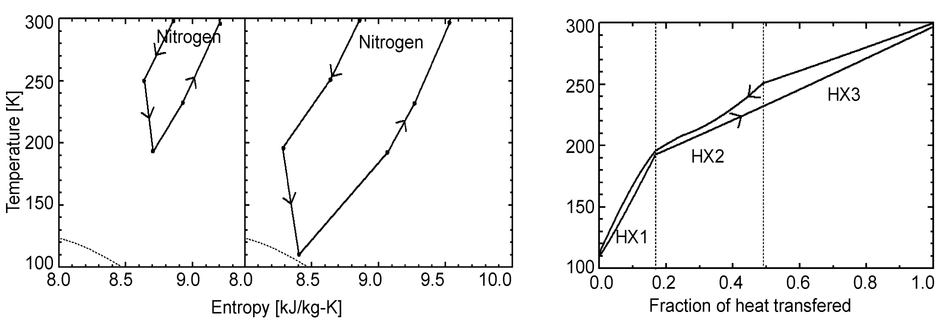
(II) Two-stage expansion cycle



(III) Dual-turbine cycle (same pressure ratio)



(IV) Dual-turbine cycle (different pressure ratio)



(IV) Dual cycle

Figure 7. Temperature-entropy diagram of nitrogen and temperature profile of hot and cold streams in heat exchangers for cycles (II)-(V)

CONCLUSIONS

Standard and modified reverse-Brayton cycles are thermodynamically investigated for the application to peak-shaving LNG processes or off-shore plants. Four different modified cycles are identified to effectively employ two turbines: two-stage expansion cycle, dual-turbine cycles with same or different pressure ratio, and dual cycle. In addition, the effect of optional pre-cooling JT cycle is considered to improve the thermodynamic efficiency. Refrigerant is nitrogen gas (N2) for all turbine cycles, and various substances for pre-cooling cycles. The cycles are simulated with commercial software on selected conditions and compared in terms of FOM and estimated size of heat exchangers.

The thermodynamic performance of standard cycle with a turbine can be greatly improved by employing two turbines in all four ways of modification. On the other hand, two-stage expansion cycle and dual-turbine cycle with same pressure ratio are preferred if the size or weight of liquefaction system is also considered. The addition of typical pre-cooling cycles can also increase the FOM of liquefaction, but the margins are not as great as the modification of cycles with two turbines. The JT cycle with ammonia (NH_3) may be recommended as a pre-cooling option, if compactness and small inventory of hydrocarbon are pursued.

ACKNOWLEDGMENT

This research is supported by a grant from the LNG Plant R&D Center funded by the Ministry of Land, Transportation and Maritime Affairs (MLTM) of the Korean government.

REFERENCES

1. Flynn, T.M., *Cryogenic Engineering, 2nd Ed.*, CRC Press, New York (2005), pp. 748-756.
2. Barron, R.F., Cryogenic System, 2nd Ed., Oxford University Press, New York (1985), pp. 60-64.
3. Venkatarathnam, G., *Cryogenic Mixed Refrigerant Processes*, Springer, New York (2008), pp. 143-199.
4. Chang, H.M., Chung, M.J., Lee, S., Choe, K.H., "An efficient multi-stage Brayton-JT cycle for liquefaction of natural gas," *Cryogenics*, Vol. 51, Issue: 6 (June 2011), pp.278-286.
5. Chang, H.M., Park, J.H., Woo, H.S., Lee, S., Choe, K.H., "New Concept of natural gas liquefaction cycle with combined refrigerants," Proceedings of Twenty-Third International Cryogenic Engineering Conference and International Cryogenic Materials Conference 2010, Oficyna Wydawnicza Politechniki Wrocławskiej, Wrocław (2011), pp. 257-262.
6. Chang, H.M., Park, J.H., Lee, S., Choe, K.H., "Combined Brayton-JT cycles with refrigerants for natural gas liquefaction," *Adv. in Cryogenic Engineering*, Vol. 57, Amer. Institute of Physics, Melville, NY (2012), pp. 1779-1788.
7. Bejan, A., *Advanced Engineering Thermodynamics, 3rd Ed.,* John Wiley & Sons, New Jersey (2006), pp. 101-142.