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New concept of natural gas liquefaction cycle with combined refrigerants

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A new concept of combined JT refrigeration cycle is presented for liquefaction process of natural gas. Parallel combnaition of two or more single-component JT cycles may have a similar thermodynamic efficiency with mixed refrigerant (MR) cycle, but still takes advantage of easy and robust operation with pure refrigerants. It is specifically demonstrated with Aspen HYSYS simulator that combined ethane and butane cycle could form an efficient pre-cooler between ambient temperature and 240 K in comparison with the reputed C3-MR and DMR processes. A patent application is underway.

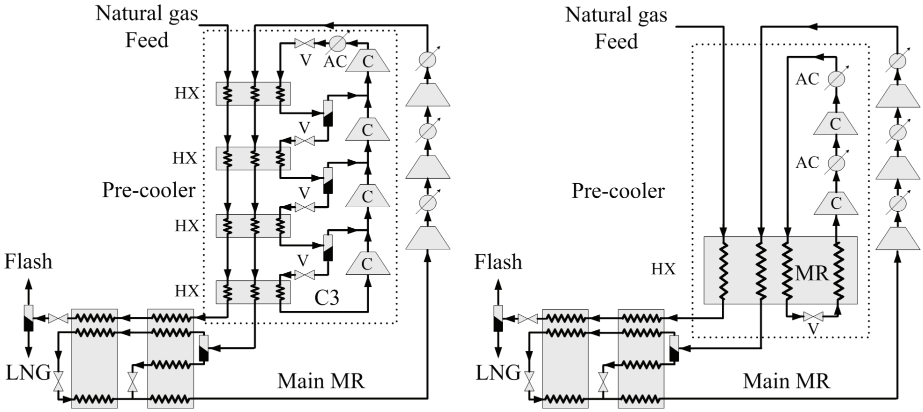
INTRODUTION

A variety of thermodynamic cycles have been developed for liquefaction of natural gas in order to meet the requirements on high efficiency, large capacity, and simple equipment [1-5]. Since natural gas is a mixture of different hydrocarbons, its enthalpy varies nonlinearly with temperature along the liquefaction process. For a high efficiency, it is important to reduce the entropy generation due to temperature difference in heat exchangers. Mixed refrigerant (MR) cycles are generally effective in reducing the temperature difference with a small number of components [4]. On the other hand, single-component refrigerant cycles are easy and robust in operation, but require a large number of refrigeration stages [2].

The most popular liquefaction process under operation is based on propane pre-cooled MR (C3-MR) cycle, as shown in Figure 1(a). Feed gas is pre-cooled to approximately 240 K by propane (C3) cycle, and then condensed and sub-cooled to 113 K by MR cycle. An appropriate composition of MR (typically, mixture of nitrogen, methane, ethane, and propane) with a phase separator allows a high thermodynamic efficiency with small number of equipments [4]. For pre-cooling, however, propane is evaporated at three or four pressure levels, depending on the ambient temperature condition.

The number of pre-cooling stages may be reduced, if another MR cycle is used. Figure 1(b) shows a dual mixed refrigerant (DMR) process, where a simple MR cycle without phase separator is the pre-cooler. The optimal composition of pre-cooling MR cycle is nearly half and half of ethane (C2) and butane (C4), and the efficiency of this DMR process is slightly lower than that of C3-MR [4].

A new concept of pre-cooling cycle is proposed in this paper to pursue the efficiency and simplicity at the same time. The key idea is to employ a parallel combination of ethane and butane JT cycles at the pre-cooling stage, as shown Figure 2. This process will be called combined refrigerant pre-cooled MR (CR-MR) process. Since the main MR cycle is same in C3-MR, DMR, and CR-MR processes, the pre-cooling cycle will determine the overall liquefaction performance.



(a) Propane pre-cooled MR (C3-MR)

(b) Dual MR (DMR)

Figure 1. Existing mixed refrigerant (MR) processes

As a first step of feasibility study, the proposed CR-MR cycle is simulated on the same basis and assumptions presented in [4], since full details of optimized cycles for C3-MR and DMR processes are available for comparison. Commercial software, Aspen HYSYS [6], is used for simulation. The optimal condition on flow rates and pressure levels is sought for the ethane and butane JT cycles in CR-MR process. The simulation results are compared with the reputed cycles in terms of thermodynamic efficiency and required number of components.

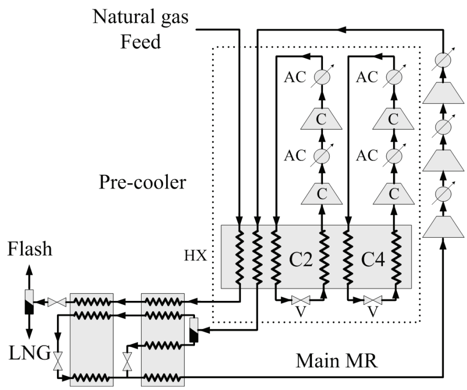


Figure 2. Newly proposed combined refrigerant (CR) pre-cooled MR processes (CR-MR)

CYCLE SIUMULATION

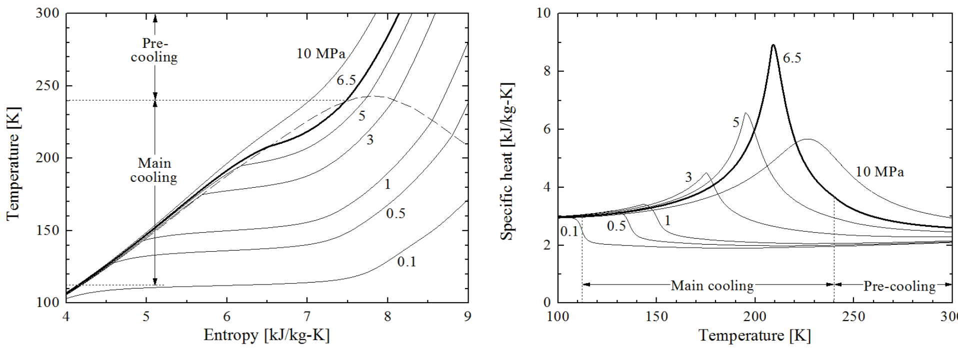
The thermodynamic design of liquefaction cycle depends to an extent on the composition, pressure, and temperature of natural gas feed. For comparison purpose in this paper, the composition of feed gas is taken from [4] and listed in Table 1. The feed gas needs to be cooled from 300 K (ambient temperature) to 113 K at 6.5 MPa for flash expansion to LNG storage.

Several isobars are plotted on temperature-entropy and specific heat-temperature diagrams in Figure 3 (a) and (b), respectively. The 6.5 MPa isobar passes near the “pseudo” critical point around at 240 K, and has a sharp peak value of specific heat around at 210 K. The pre-cooling temperature is assumed to be 240 K, and the optimized compositions of main MR and pre-cooling MR are also listed in Table 1. In addition, the following assumptions are made for simplicity [4].

1. The pressure drop in all heat exchangers and phase separators is zero.
2. The minimum temperature approach between the hot and cold streams in all heat exchangers is 3 K.
3. The adiabatic efficiency of all compressors is 80%.
4. The temperature of after-cooling refrigerant is 315 K for C3 and 310 K for MR and CR.

Table 1. Composition of natural gas feed and refrigerant [4]

Feed and Refrigerant (Process)		Component (mole %)						
		Nitrogen	Methane	Ethane	Propane	nButane	iButane	iPentane
Natural Gas Feed (All processes)		4.0	87.5	5.5	2.1	0.5	0.3	0.1
Main MR (All processes)		7.0	41.8	29.9	21.3	-	-	-
Precooling Refrigerant	Propane (C3-MR)				100			
	MR (DMR)			45.5	4.9	49.6		
	Combined (CR-MR)			100		100		



(a) Isobars on temperature-entropy diagram

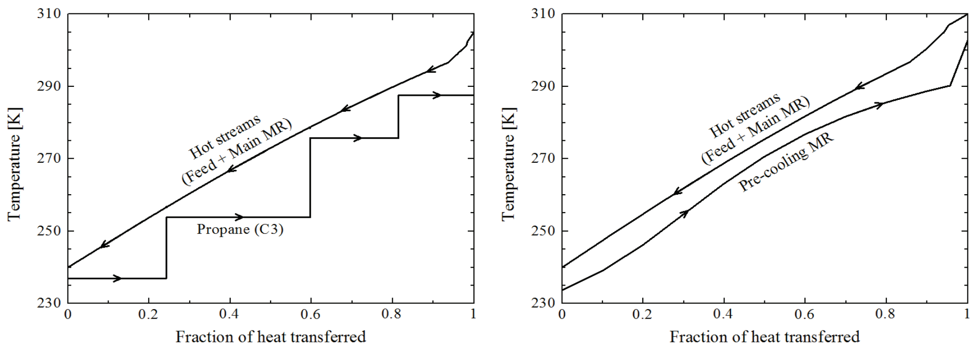
(b) Specific heat as a function of temperature

Figure 3. Thermodynamic properties of natural gas feed

RESULTS AND DISCUSSION

Figures 4(a) and (b) show the simulated temperature profiles in pre-cooling heat exchangers of C3-MR and DMR processes, respectively. Since propane (C3) is a pure refrigerant, the temperature profile is stepwise in Figure 4(a), and the minimum difference (3 K) occurs at every step. On the contrary, the temperature profile of MR varies gradually in Figure 4(b), and the minimum difference (3 K) at the middle of heat exchanger. The pressure levels of propane cycle and the composition of pre-cooling MR cycle are optimized, as noted in [4].

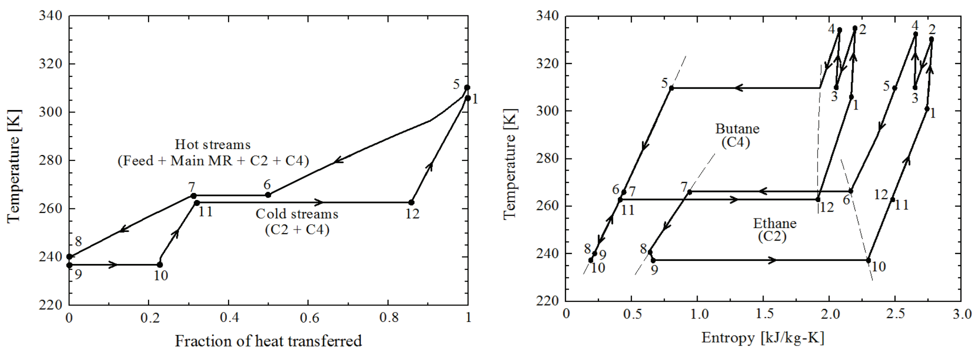
Figure 5(a) is the simulated temperature profile in pre-cooling heat exchanger for the combined refrigerant (CR) cycle with ethane (C2) and butane (C4). For better explanation, the temperature-entropy diagram of ethane and butane cycles is plotted in Figure 5(b). Since two cold refrigerants enter the heat exchanger as liquid at 9, the temperature of cold streams has two horizontal regions (9-10 and 11-12) corresponding to evaporation of C2 and C4, respectively. On the other hand, the temperature of hot streams has only one horizontal region (6-7) corresponding to condensation of C2, because C4 enters as liquid at 5.



(a) C3-MR process

(b) DMR process

Figure 4. Temperature profiles of hot and cold streams in pre-cooling heat exchangers



(a) Temperature profiles of hot and cold streams

(b) Temperature-entropy diagram

Figure 5. Simulation results of pre-cooling process with CR (C2+C4)

The flow rate and pressure levels of two parallel cycles should be determined to narrow the temperature difference or to achieve a high thermodynamic efficiency. The optimization method is more or less similar to that to search the optimal composition and pressure levels of MR cycles [4]. Figure 5 is the best result that the present authors have obtained so far. It is noted that the temperature difference is 3 K at the cold and warm ends, and the evaporation temperature of C4 is also 3 K higher than the condensation temperature of C2. In this case, the C4 cycle plays a role of topping cycle for C2 cycle (as in cascade), even though the two cycles are combined in parallel.

The exergy balance in pre-cooler [4] can be written as

$$\sum \dot{W}_C = \dot{m}_F\left[\left(h_e - h_i\right) - T_0\left(s_e - s_i\right)\right]_F + \dot{m}_{MR}\left[\left(h_e - h_i\right) - T_0\left(s_e - s_i\right)\right]_{MR} + T_0 \sum \dot{S}_{gen} \tag{1}$$

where the left-handed side is the total power input to the pre-cooling refrigerants, and the next two bracket terms are the useful effect (i.e. the exergy increase) of feed gas (F) and main MR, respectively. The difference between power input and exergy increase is so-called irreversibility, which is the entropy generation rate multiplied by ambient temperature. The contribution of entropy generation can be itemized for all the components in the pre-cooler.

Figure 6 shows the exergy utilization and irreversibility in the pre-cooler for the three processes. The exergy efficiency (defined as the ratio of exergy increase to the power input) is 34.3% and 30.5% for the pre-cooler of C3-MR and DMR processes, respectively, which are in good agreement with [4]. The exergy efficiency of CR pre-cooler is estimated at 31.5% on the same basis and assumptions. It is recalled that the CR cycle has only one stage of parallel heat exchanger, while the C3 cycle has four refrigeration stages in series. It can be also mentioned that the CR cycle has a slightly higher efficiency than the pre-cooling MR cycle, but still takes advantage of pure refrigerants in easy and robust operation.

The contribution of irreversibility in each cycle is subdivided into four groups: valves (V), after-coolers (AC), compressors (C), and heat exchangers (HX), as shown in Figure 6. It may be noted that four valves are responsible for relatively large irreversibility in C3-MR, and that the after-coolers are the main source of inefficiency in DMR. In the proposed CR-MR, on the contrary, the heat exchanger generates more irreversibility than the valves or after-coolers, when compared with the two reputed processes.

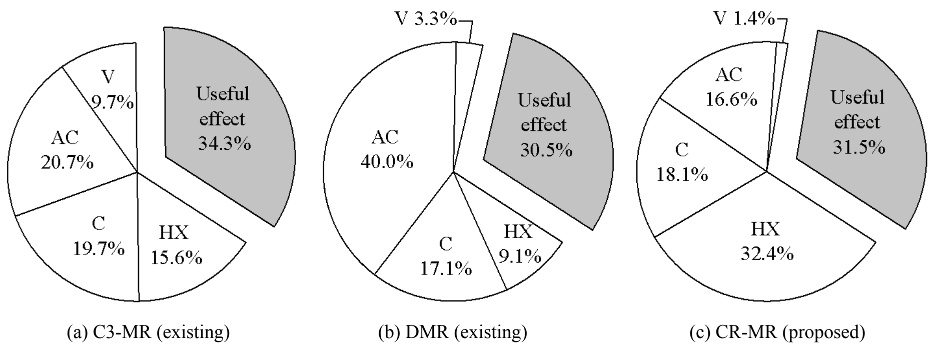


Figure 6 Exergy utilization of pre-cooler for existing and proposed processes
(Useful effect: exergy increase, V: valves, AC: after-coolers, C: compressors, HX: heat exchangers)

In practice, the actual performance of CR pre-cooler may be somewhat different from the simulated results, because the six (two cold and four hot) streams in a heat exchange may be coupled each other in various ways. The presented simulation results are just useful in demonstrating the feasibility of the proposed CR-MR process, as the required energy balance is satisfied with the constraint of minimum temperature difference.

CONCLUSIONS

Parallel combination of ethane (C2) and butane (C4) JT cycles is proposed as a pre-cooler of main MR cycle for natural gas liquefaction. To investigate the feasibility, the pre-cooling cycle is simulated with Aspen HYSYS, and the results are compared with the reputed C3-MR and DMR processes. It may be concluded that the pre-cooler with combined refrigerant (CR) can take advantages of high efficiency and simple hardware at the same time. Towards practical development, a patent application is underway, and further studies are recommended for the optimal operating conditions and the detailed performance of multiple-passage heat exchangers.

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