Research paper

Thermodynamic design of hydrogen liquefaction systems with helium or neon Brayton refrigerator

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**A B S T R A C T**

A thermodynamic study is carried out for the design of hydrogen liquefaction systems with helium (He) or neon (Ne) Brayton refrigerator. This effort is motivated by our immediate goal to develop a small-capacity (100 L/h) liquefier for domestic use in Korea. Eight different cycles are proposed and their thermodynamic performance is investigated in comparison with the existing liquefaction systems. The proposed cycles include the standard and modified versions of He Brayton refrigerators whose lowest temperature is below 20 K. The Brayton refrigerator in direct thermal contact with the hydrogen flow at atmospheric pressure from ambient-temperature gas to cryogenic liquid. The Linde-Hampson system pre-cooled by a Ne Brayton refrigerator is also considered. Full cycle analysis is performed with the real properties of fluids to estimate the figure of merit (FOM) under an optimized operation condition. It is concluded that He Brayton refrigerators are feasible for this small-scale liquefaction, because a reasonably high efficiency can be achieved with simple and safe (low-pressure) operation. The complete cycles with He Brayton refrigerator are presented for the development of a prototype, including the ortho-to-para conversion.

**1. Introduction**

Hydrogen liquefaction is an important thermodynamic process that has been fully developed for large-scale cryogenic applications [1,2]. Over decades, a notable quantity of liquid hydrogen has been consumed as the propellant of rocket engines and space launch vehicles [2]. The hydrogen bubble chamber uses liquid hydrogen in the detection and study of high-energy particles [1]. The spallation neutron source also uses liquid hydrogen for removing the huge amount of dissipated energy from neutron moderators at 20 K or lower temperatures [3]. Recently, new large-scale applications have been proposed and explored, such as a hybrid energy transfer of liquid hydrogen through superconducting power cables [4], and an ocean-going transport of liquid hydrogen for international energy trade [5].

In order to meet these needs, the hydrogen liquefiers with capacity of 500–3000 L/h are supplied by major gas companies [6,7]. Since the large-scale system has been installed at some limited locations, liquid hydrogen may not be locally available for smaller-scale application in many other regions. In Korea, for example, liquid hydrogen is not commercially available, although a potential market is now emerging for the car or truck fueling station [8] and the power package of unmanned air vehicles or drones [9].

This thermodynamic study is motivated by our immediate goal to design and construct a 100 L/h liquefier for domestic use in Korea. The most suitable thermodynamic cycle for smaller-capacity liquefaction may be different from that of the full-capacity liquefiers, taking into consideration not only the energy efficiency and economic factors, but also the practical issues like the safety and simplicity in operation. As a beginning step of the development program, a variety of refrigeration cycles for liquefaction are proposed and their feasibility is investigated in comparison with the existing liquefaction systems.

Fig. 1 compares schematically the thermodynamic structure of refrigerator, liquefier, and refrigerator for liquefaction [10]. A refrigerator operates in closed cycle, receiving the thermal load at cryogenic temperature and rejecting the heat to ambient. On the other hand, a liquefier operates in open cycle, where gas is fed at ambient temperature and liquid is delivered at cryogenic temperature. In most cases, the feed gas itself is the working fluid that undergoes compression and expansion. A refrigerator for liquefaction operates in closed cycle, but the thermal load is distributed over the liquefaction stream from gas at ambient temperature to liquid at cryogenic temperature.

There are various options in selecting the refrigeration cycle for hydrogen liquefaction, as far as the cold temperature is lower than 20 K. A simple and convenient method is to employ a Gifford-McMahon (GM)
or pulse tube cryocooler, whose cold-head temperature is below 20 K. It was recently reported that a small rate of liquefaction (1 L/h) was achieved with a single-stage GM cooler and liquid-nitrogen (LN₂) pre-cooling [11]. Because of the limit in refrigeration capacity, these regenerative cryocoolers are not virtually applicable to 100 L/h liquefaction.

A dominant choice is to use a helium (He) Brayton refrigerator, in the similar way as nitrogen (N₂) Brayton refrigerators are widely utilized for the liquefaction of natural gas [10] or methane [12]. Recently, Chang et al. [13] published a paper on standard or modified versions of He Brayton refrigeration cycle for liquid hydrogen below 20 K in the neutron moderators under construction at European Spallation Source (ESS). It was reported that the thermodynamic performance of standard Brayton cycle could be significantly improved by employing two turbo-expanders in series or in parallel. The similar modifications could be effective in hydrogen liquefaction as well. Another choice is to use a neon (Ne) Brayton refrigerator as pre-cooler of Linde-Hampson system. Since the normal boiling temperature of Ne is 27 K, the Ne Brayton refrigerator is not capable of liquefying hydrogen by itself, but may be useful for pre-cooling the hydrogen flow to Joule-Thomson (JT) valve. Lately, a major gas company has successfully developed Ne Brayton refrigerators with a capacity of 2–10 kW at 60–70 K [14,15], which could be shortly modified for this application. The objective of this thermodynamic study is to identify the most feasible cycles for small-scale hydrogen liquefaction and determine the key parameters of a 100 L/h liquefier for prototype construction.

2. Existing and proposed cycles

2.1. Existing cycles

The Linde-Hampson system is obviously desirable for small-scale liquefaction, because of its simplicity. For hydrogen liquefaction, however, a pre-cooling is required, because the maximum inversion temperature is lower than ambient temperature [1]. The LN₂ pre-cooled Linde-Hampson system for hydrogen liquefaction is shown in Fig. 2(a). In general, this system requires a very high pressure of hydrogen, because the production of cryogenic liquid relies only on the Joule-Thomson (JT) process. It is noted that the LN₂ heat exchanger (HX1) in Fig. 2(a) has three streams.

Many industrial hydrogen liquefiers are based on Claude cycle [1]. Fig. 2(b) shows the standard Claude system for hydrogen liquefaction. The high-pressure gas is diverted from the main stream, expanded through a turbo-expander (E), and reunited with the low-pressure stream. The stream to be liquefied continues to a JT valve at the cold end. The turbo-expander is a key component, where the adiabatic expansion is effectively used for the production of low temperature. Fig. 2(c) shows the Claude system with LN₂ pre-cooling.

2.2. Proposed cycles

Standard and modified He Brayton cycles are proposed for hydrogen liquefaction. The standard He Brayton refrigerator is shown in Fig. 2(d), and the He Brayton refrigerator with LN₂ pre-cooling is shown in Fig. 2(e). While the Linde-Hampson and Claude systems are classified as a liquefier in Fig. 1, the He Brayton systems are classified as a refrigerator for liquefaction. The lowest temperature of He gas at the exit of turbo-expander (E) must be lower than 20 K, and the liquefaction flow of hydrogen may be at atmospheric pressure. It is noted that the HG’s have multiple (three or four) streams in these systems.

In order to improve the thermodynamic performance, the He Brayton cycle is modified by employing two turbo-expanders in different ways [13]. Fig. 2(f) shows 2-stage He Brayton refrigerator, where two expanders are arranged in series. Alternatively, Fig. 2(h) shows dual-turbine He Brayton refrigerator, where two expanders are arranged in parallel. Fig. 2(g) and (i) show the 2-stage He refrigerator with LN₂ pre-cooling and the dual-turbine He refrigerator with LN₂ pre-cooling, respectively. In 2-stage systems, two turbines have the same flow rate, but different pressure ratios. In dual-turbine systems, on the contrary, two turbines have the same pressure ratio, but different flow rates.

Fig. 2(j) shows the Linde-Hampson system pre-cooled by Ne Brayton refrigerator, and Fig. 2(k) shows the Linde-Hampson system pre-cooled by LN₂ and Ne Brayton refrigerator. These systems are more complicated in a sense that two separate cycles (Ne refrigeration cycle and H₂ liquefaction cycle) are combined.