Thermodynamic Analysis of Hydrogen Liquefaction Systems Using Gifford-McMahon Cryocooler

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Key words : Liquefaction, Hydrogen, Liquid, Optimum design, GM cryocooler, cryogenics

Abstract

Thermodynamic cycle analysis is presented to estimate the maximum liquefaction rate of hydrogen for various systems using a Gifford-McMahon(GM) cryocooler. Since the present authors' previous experiments showed that the gaseous hydrogen was liquefied approximately at the rate of 5.1 mg/s from the direct contact with a commercial two-stage GM refrigerator, this study has been proposed to predict how much the liquefaction rate can be increased in different configurations using the GM cooler and with improved heat exchangers. The optimal operating conditions have been analytically sought with real properties of normal hydrogen for the Linde-Hampson(L-H) system precooled by single-stage GM, the direct-contact system with two-stage GM, the L-H system precooled by two-stage GM, and the direct-contact system with helium GM-JT (Joule-Thomson). The maximum liquefaction rate has been predicted to be only about 7 times greater than the previous experiment, even though the highly effective heat exchangers may be employed. It is concluded that the liquefaction rate is limited mainly because of the cooling capacity of the commercially available GM cryocoolers and a practical scale of hydrogen liquefaction is possible only if the GM cooler has a greater capacity at 70-100 K.

Nomenclature

h : specific enthalpy [J/g]

$\dot{m}$: mass flow rate [g/s]

P_H : high pressure [bar]

P_L : low pressure [bar]

$\dot{Q}$: heat transfer rate [W]

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T : temperature [K]
$\dot{W}$: power input [W]
y : liquid yield

Greek symbols

ε : effectiveness of heat exchanger

Subscripts

C : compressor
f : liquid phase
g : vapor phase
ref : refrigerator or GM cryocooler
H_2 : hydrogen
H_e : helium
PC : precooling
0,1,2,.. : location in liquefier

1. Introduction

Liquefaction of hydrogen at cryogenic temperatures is an excellent method of storage, as fuel since the weight or the volume per unit stored energy is much lower than the other methods such as the compressed gas or the metal hydride.[1,2] At the beginning stage of the cryogenic engineering, hydrogen was liquefied by the conventional cascade refrigeration, but it has not been utilized in commercial scale because of its complexity and poor thermodynamic efficiency. In most of the hydrogen plants built in mid 20th century or later, the liquefaction is based upon Claude cycle[3].

In the Claude systems, a cryogenic expander is the key component to produce an external work from the compressed gas. The technology to design and manufacture the reciprocating or turbo expanders has been developed exclusively in the U.S. and a few European nations, and can not be domestically realized in near future. It has been lately proposed to develop a small-scale liquefaction system without the cryogenic expanders. The system may be useful in local liquefaction at the automobile fueling stations, in order to eliminate the thermal loss for the storage and the transfer of the cryogenic liquid.[8]

A simple small-scale system is possible by direct-contact[9] of the hydrogen gas with a cryocooler whose surface temperature is lower than the boiling temperature or by the Linde-Hampson system precooled by a cryocooler. Recently Kim et. al[10,11] presented basic design data for such systems and later reported the optimal conditions in the Linde-Hampson system precooled by some commercial Gifford-McMahon(GM) coolers. Baik et. al[12] successfully liquefied hydrogen by the direct-contact method with a two-stage GM cryocooler.

This study is focused specifically on the maximum liquefaction rate in the various systems for a given GM cryocooler. The energy efficiency of the liquefaction is outside of our interest in this paper. In each system, a detailed thermodynamic cycle analysis is performed to calculate the maximum rate, with the cooling capacity data of the GM cooler provided by the manufacturer. For a particular case that the experimental data are available, the analysis results are compared with the corresponding data. It is finally intended to predict how much the liquefaction rate can be practically obtained by the several GM coolers in the market.

2. Analysis model

A simple Linde-Hampson(L-H) liquefaction system is schematically shown in Fig. 1(a). The hydrogen gas at room temperature is compressed from the atmospheric pressure to a

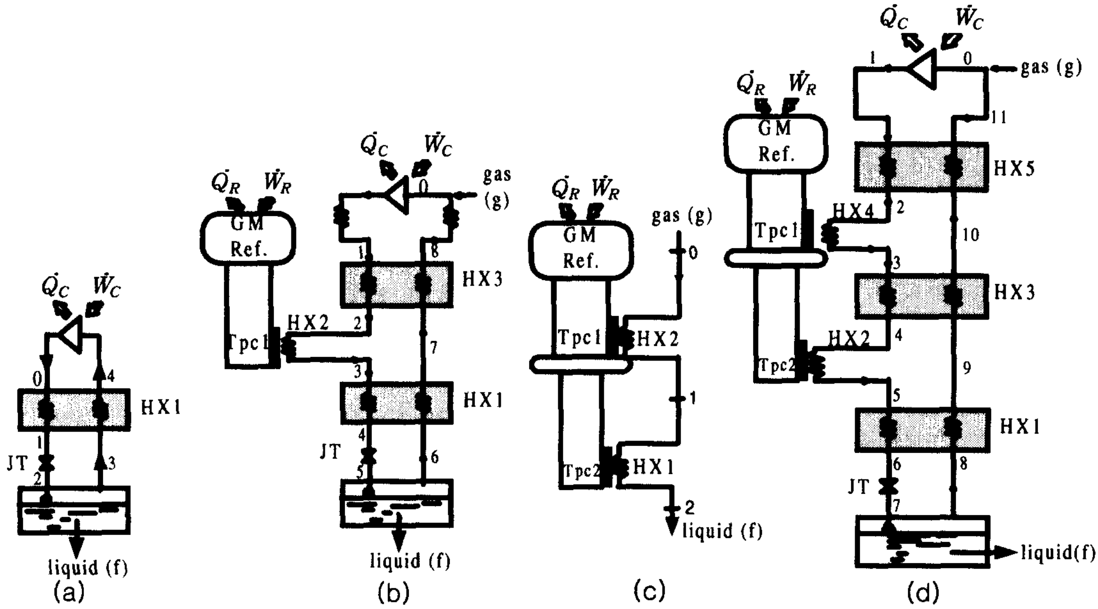

Fig. 1 Schematic diagram of hydrogen liquefaction systems using GM cryocooler (a) Simple L-H system (b) L-H system precooled by single-stage GM (c) Direct-contact system with two-stage GM (d) L-H system precooled by two-stage GM.

high pressure at the compressor and then cooled through a counterflow heat exchange with the cold vapor stream from the flash chamber. The cooled high-pressure gas is expanded to the atmospheric pressure through Joule-Thomson(JT) valve, exiting in a saturated state having the lowest temperature. Only the saturated liquid is extracted from the bottom of the flash chamber and the saturated vaper from the top is returned to the heat exchanger.

Since the maximum inversion temperature[3] of hydrogen is lower than the room temperature, hydrogen can not be liquefied in the simple L-H system. The L-H system should be modified by "precooling" so that the warm end temperature of the heat exchanger may be lower than the inversion temperature. In this paper, the four different configurations are considered, including the L-H systems precooled by a single-stage or two-stage GM, the direct-contact systems, and the helium GM-JT systems.

2.1 L-H System precooled by single-stage GM

Figure 1(b) shows the Linde-Hampson system which is precooled by a single-stage GM cooler. It is composed of a compressor, a GM cooler, two counterflow heat exchangers, a JT valve and a flash chamber. The cold-head temperature of the GM cryocooler should be well below the maximum inversion temperature of hydrogen.

The energy balance equations for HX3 and HX2 can be written in terms of the specific enthalpy at the inlets and the exits.

$$h_1 - h_2 = (1-y)\cdot(h_8 - h_7) \tag{1}$$

$$\dot{Q}_{1ref} = \dot{m}(h_2 - h_3) \tag{2}$$

where y is the liquid yield and $\dot{m}$ is the mass flow rate at the compressor. Since the work and the heat transfer are assumed to be zero at JT valve, the energy balance is simply reduced to

$$h_4 = h_5 \tag{3}$$

From the energy balance for the insulated flash chamber at the cold end, the liquid yield and the liquefaction rate ($\dot{m}_f$) are related as following,

$$y = \frac{(h_6 - h_4)}{(h_6 - h_f)} = \frac{\dot{m}_f}{\dot{m}} \tag{4}$$

The effectiveness of the heat exchangers should be expressed in terms of enthalpy, instead of temperature, because the specific heat of the gas is a strong function of temperature at these cryogenic conditions. For example, if the high-pressure stream has a heat capacity greater than the low-pressure side or

$$h_1 - h(T_7, P_H) \geq (1-y)\cdot[h(T_1, P_L) - h_7] \tag{5}$$

the effectiveness of HX3 is given as

$$\varepsilon_3 = \frac{h_8 - h_7}{h(T_1, P_L) - h_7} \tag{6}$$

Otherwise, it is given by

$$\varepsilon_3 = \frac{h_1 - h_2}{h_1 - h(T_7, P_H)} \tag{7}$$

In case of HX2 which is in contact with the refrigerator, the effectiveness is expressed as

$$\varepsilon_2 = \frac{h_2 - h_3}{h_2 - h(T_{PC1}, P_H)} \tag{8}$$

Most of the manufacturers of the GM coolers provide the cooling capacity as a function of the refrigeration temperature.

$$\dot{Q}_{1ref} = f(T_{PC1}) \tag{9}$$

In this analysis, we have 9 unknowns, including the temperatures at each location, the liquid yield and the cooling capacity of the cooler. ($T_2, T_3, T_4, T_5, T_7, T_8, T_{PC1}, y, \dot{Q}_{1ref}$) The 9 equations to determine these unknowns are constituted by the five energy balance equations, the three effectiveness equations for the HX's and the cooling capacity equation. Once the input variables are given for the high pressure, the mass flow rate and the effectiveness of the heat exchangers, the equations are solved simultaneously with an iterative method to calculate the liquefaction rate.[14,15]

2.2 Direct contact system with two-stage GM

One of the simplest liquefaction methods is to keep the hydrogen gas in a thermal contact with a refrigerator whose surface temperature is below the boiling temperature as shown in Fig. 1(c). Most of the commercial two-stage GM cryocoolers reach well below 20 K, they can be used for the liquefaction of hydrogen.

The cooling capacity of the two-stage cooler is more complicate than that of the single-stage cooler. In general, the cooling capacity at each stage is expressed as a function of the first stage and the second stage temperatures. The energy balance equations can be written as

$$\dot{Q}_{1ref} = f(T_{PC1}, T_{PC2}) = \dot{m}_f(h_0 - h_1) \tag{10}$$

$$\dot{Q}_{2ref} = g(T_{PC1}, T_{PC2}) = \dot{m}_f(h_1 - h_2) \tag{11}$$

The effectiveness of the two heat exchangers, HX1 and HX2, are expressed as respectively.

$\dot{Q}_C$ $\dot{W}_C$ $\dot{Q}_R$ $\dot{W}_R$

$$\varepsilon_1 = \frac{h_1 - h_2}{h_1 - h(T_{PC2}, P_L)} \quad (12)$$

$$\varepsilon_2 = \frac{h_0 - h_1}{h_0 - h(T_{PC1}, P_L)} \quad (13)$$

In the analysis of this system, we have six unknowns, including the temperature at state 1, the mass flow rate, the temperature and the cooling capacity at the two stages. The six equations given by Eq. (10)-(13) are solved simultaneously to determine the mass flow rate, which is also the liquefaction rate.[15]

2.3 L-H System precooled by two-stage GM

The analysis for the Linde-Hampson system precooled by a two-stage GM cooler is basically the same as that described in Section 2.1, except that there are two precooling stages as shown in Fig. 1(d). The cooling capacity at each stage is expressed by the two precooling temperatures, so the energy balance equations are.

$$\dot{Q}_{1ref} = f(T_{PC1}, T_{PC2}) = \dot{m}(h_2 - h_3) \quad (14)$$

$$\dot{Q}_{2ref} = g(T_{PC1}, T_{PC2}) = \dot{m}(h_4 - h_5) \quad (15)$$

The effectiveness of HX1, HX3 or HX5 is given in the same way as Eq. (6) or (7), and the effectiveness of HX2 or HX4 is given in the same way as Eq. (8).

In this analysis, we have 14 unknowns, including T_2, T_3, T_4, T_5, T_6, T_7, T_9, T_{10}, T_{11}, T_{PC1}, T_{PC2}, $\dot{Q}_{1ref}$, $\dot{Q}_{2ref}$, and y. The 14 equations are derived from the energy balance for the 7 components (5 heat exchangers, a JT valve, a flash chamber), the effectiveness relation for the 5 heat exchangers, and the two expressions for the cooling capacity at two stages. The liquefaction rate is calculated after the state at every location is determined by a proper iterative procedure.[15]

2.4 Direct contact system with helium GM-JT

Another option for the liquefaction of hydrogen using the GM cooler is to incorporate a GM-precooled Joule-Thomson circuit with gas helium, as shown Fig. 2. In this system, a two-stage GM cryocooler acts as both a precooler of helium JT circuit and a direct-cooler of hydrogen. The hydrogen gas cooled by the direct contact with the two stages of the GM cooler is finally liquefied by the counterflow heat exchange (HX8) with the cold helium of the JT circuit.

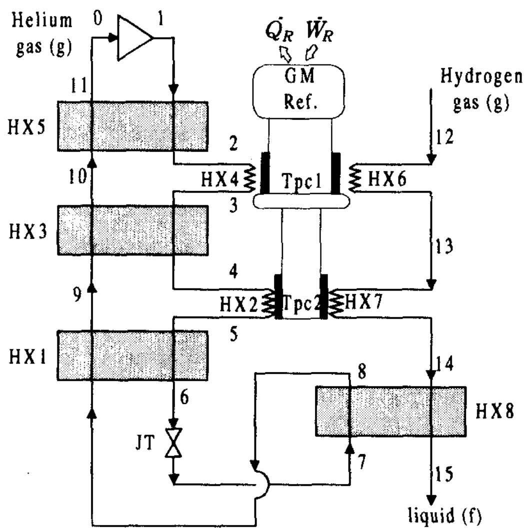

Fig. 2 Schematic diagram of contact liquefaction system with two-stage helium GM-JT refrigerator.

Because of the thermodynamic properties of hydrogen, the optimal high pressure of Linde-Hampson system is much greater than 100 bars as will be discussed in next sections. On the other hand, the closed JT cycle of helium can be operated as pressures not exceeding about 20 bars, which is the main advantage of this system.

The cycle analysis includes the procedures for the systems of both Fig. 1(c) and (d). The energy balance for the HX8 can be expressed as

$$\dot{m}_{He}(h_8-h_7)=\dot{m}_{H_2}(h_{14}-h_{15}) \tag{16}$$

The cooling capacity at each stage of the GM cooler should be the sum of the cooling of hydrogen gas and that of the helium gas.

$$\dot{Q}_{1ref}=\dot{m}_{He}(h_2-h_3)+\dot{m}_{H_2}(h_{12}-h_{13}) \tag{17}$$

$$\dot{Q}_{2ref}=\dot{m}_{He}(h_4-h_5)+\dot{m}_{H_2}(h_{13}-h_{14}) \tag{18}$$

This analysis includes have 17 unknowns, which are T_2, T_3, T_4, T_5, T_6, T_7, T_8, T_9, T_{10}, T_{11}, T_{13}, T_{14}, T_{PC1}, T_{PC2}, $\dot{Q}_{1ref}$, $\dot{Q}_{2ref}$ and $\dot{m}_{H_2}$. Among the 17 equations, seven are derived from the energy balance for each component, eight from the effectiveness relation of each heat exchanger, and two from the cooling capacity of each stage. The input variables to perform the analysis are the mass flow rate and high pressure of helium, and the effectiveness of each heat exchanger. By an iterative method, every state in the system is determined and the liquefaction rate of hydrogen is calculated.[15]

3. Results and discussion

3.1 L-H System precooled by single-stage GM

Among the commercial single-stage GM coolers to precool the Linde-Hampson system, four specific models have been selected for this analysis, AL05 of Cryomech, CGR011 of CVI, DE108 of APD, and RGS273 of Leybold.

Thermodynamic properties of hydrogen are calculated by the commercial program developed by the NIST.[13] Hydrogen is assumed to be normal hydrogen for simplicity. The effect of the conversion from ortho-to para-hydrogen on the liquefaction is treated in details in the project report.[15]

For a single-stage GM cooler, one of the nice fitting curves can be expressed as

$$\dot{Q}_{1ref}=A+B\log(T_1) \tag{19}$$

where the constants A and B are determined from the performance data provided by the manufacturer. Figure 3 shows the performance curves for the four selected coolers. Figure 4 shows the liquefaction rate as a function of high pressure for various mass flow rates, in case that the GM cooler is Cryomech Model AL05 and the heat exchanger effectiveness is 93%. For a given mass flow rate, the liquefaction

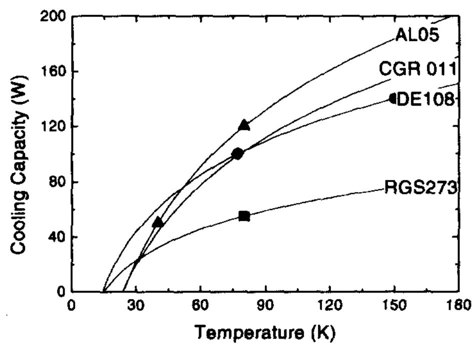

Fig. 3 Cooling capacity as a function of temperature for various single-stage GM cryocoolers.

rate increases to a maximum and then decreases as the high pressure increases.

The optimum high pressure for the maximum liquefaction is around 125 bars. It is also observed that as the mass flow rate increases, the liquefaction rate increases to a maximum and then decreases, so the optimal mass flow is approximately 140 mg/s in this specific case.

It can be generally stated that there exist unique optimal values for the mass flow and the high pressure to maximize the liquefaction rate. The existence of the optimum high pressure is related with the thermodynamic properties of hydrogen, while that of the optimum mass flow rate is due to the limited cooling capacity of the GM cooler. Figure 5 shows the maximum liquefaction rate as a function of the mass flow rate when the high pressure is optimized and the effectiveness of heat exchangers is 93%. The same procedures have been repeated for various values of the HX effectiveness and the results are shown in Fig. 6. As the effectiveness increases, the liquefaction rate increases and the corresponding optimum for the mass flow rate also increases.

The optimal conditions and the maximum liquefaction rate are listed in Table 1 for various single-stage GM coolers. The optimum cooling temperature of the GM is around 60-70 K when $\varepsilon = 0.93$, and 90-100 K when $\varepsilon = 0.99$. It is obvious that the GM cooler should have a large capacity at this precooling temperatures to be useful in this Linde-Hampson system.

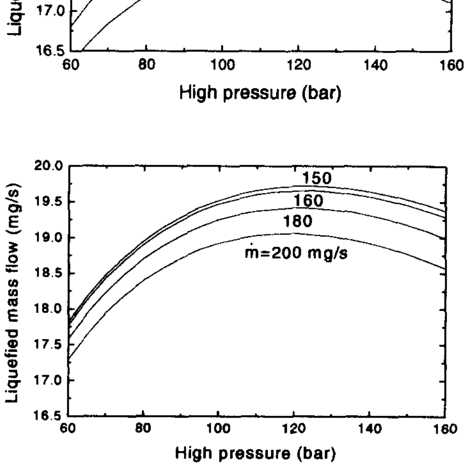

Fig. 4 Effect of high pressure on the liquefaction rate for various mass flow rates in system of Fig.1(b), when ε =0.93.

3.2 Direct contact system with two-stage GM

A number of two-stage GM cryocoolers can be found in market, such as Models 1020CP and 1050CP of CTI, GB07 and GB37 of Cryomech, DE 208R and DE 208L of APD. Unfortunately, however, most of the manufacturers provide

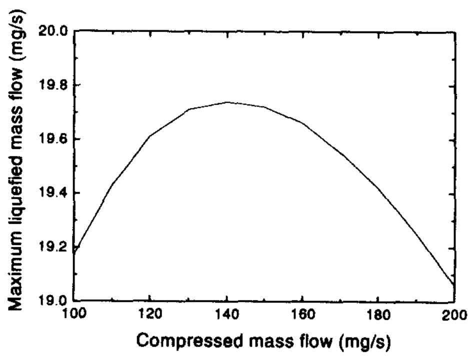

Fig. 5 Maximum liquefaction rate vs mass flow rate in system of Fig. 1(b), when ε =0.93.

Table 1 Optimal conditions and maximum liquefied mass flow for four different cryocoolers and two different HX ε's in system of Fig. 1(b)

		Cryomech AL 05		CVI CGR 011		APD DE 108		Leybold RGS 273	
HX Effectiveness: ϵ		0.93	0.99	0.93	0.99	0.93	0.99	0.93	0.99
Optimal conditions	Precooling temperature (K)	73.86	101.0	74.06	100.2	67.56	94.97	75.12	93.75
	Precooling load (W)	111.94	143.54	95.47	121.0	92.15	112.58	52.91	60.26
	High pressure (bar)	125	90	125	85	115	85	115	85
	Mass flow (mg/s)	140	420	120	360	100	290	50	150
Maximum liquefaction rate (mg/s)		19.74	28.78	16.80	24.4	17.41	23.46	9.32	12.64
Liquid yield: y		0.141	0.069	0.14	0.068	0.174	0.08	0.186	0.084

their cooling capacity only when the first and the second stages are at 77 K and 20 K, respectively. This analysis has been restricted to 1020CP and 1050CP of CTI, for which the full performance data are available.

For a two-stage GM cooler, the first stage capacity, Q_1, is generally a function of the two temperatures, T_1 and T_2, and the second stage capacity, Q_2, is also a function of T_1 and T_2, Fairly good expressions to fit the available capacity data are

$$\dot{Q}_{1ref} = A(T_2) + B(T_2)\log(T_1) \tag{20}$$

$$\dot{Q}_{2ref} = C(T_1) + D(T_1)\log(T_2) \tag{21}$$

In the liquefaction system in a direct contact with two-stage GM cooler, the mass flow rate of hydrogen, which is also the liquefaction rate, is determined by the cooling capacity at the two stages. When the effectiveness of heat exchange at each stage is 0.93, the liquefaction rate is 5.1 mg/s for Model 1020CP and 3.7 mg/s for Model 1050CP. If the effectiveness is 1.0 in its extreme limit, the liquefaction rate is predicted to be 11.6 mg/s and 11.4 mg/s, respectively for the two models. The result for Model 1020CP is in a good agreement with the experiment.[12,15] It is interesting for the two models to result in almost the same liquefaction rate as the effectiveness approaches to 1.0, since the former has a greater capacity at the second stage and the latter has a greater capacity at the first stage.

3.3 L-H System precooled by two-stage GM

From the similar physics as described in Section 3.1, there also exist the unique optimal values for the mass flow rate and the high

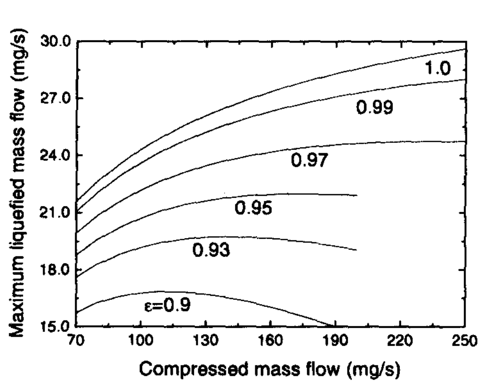

Fig. 6 Maximum liquefaction rate vs mass flow at the optimal high pressure for various ε's, in system of Fig. 1(b).

pressure to maximize the liquefaction rate in the Linde-Hampson system precooled by a two-stage GM cooler. Figure 7 shows the liquefaction rate as a function of high pressure for various mass flow rates when the GM cooler is CTI 1020CP and the effectiveness of heat exchangers is 0.93. Figure 8 shows the maximum liquefaction rate as a function of the mass flow rate when the high pressure is optimized and ε=0.93. The same procedures have been repeated for various ε's and the results are plotted in Fig. 9. Again, as the effectiveness increases, the liquefaction rate increases and the corresponding optimum for the mass flow rate also increases.

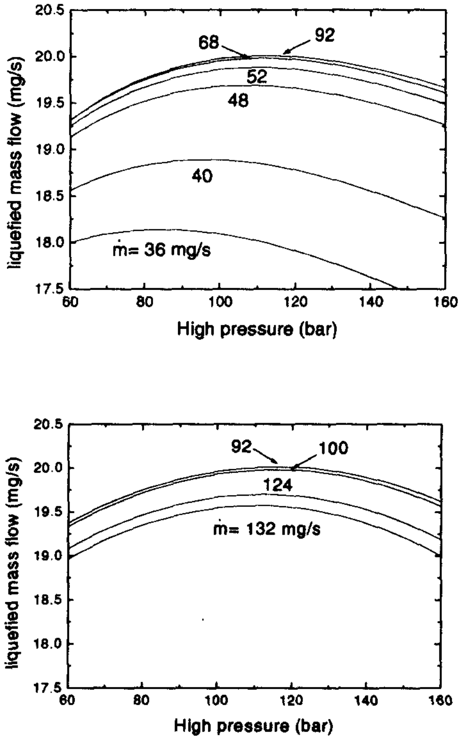

Fig. 7 Effect of high pressure on liquefaction rate for various mass flow rates in system of Fig. 1(d), with CTI 1020CP when ε=0.93.

The optimal conditions and the maximum liquefaction rates for the models 1020CP and 1050CP of CTI are summarized in Table 2. As noted by footnote, the direct-contact experiment with 1020CP has produced about 5.1 mg/s of liquefaction. It is predicted that the liquefaction of direct-contact can be merely doubled by maximizing the heat exchange between the cold heads of the GM cooler and the hydrogen gas. On the other hand, the liquefaction may be increased approximately by 4 or 5 times, depending on the heat exchanger effectiveness, if the 1020CP is used as a precooler of the Linde-Hampson system. Since the Model 1050CP has a greater capacity at the first stage and a less capacity at the second stage, a smaller liquefaction rate is predicted for direct-contact system while a larger rate is predicted for the precooled Linde-Hampson system.

3.4 Direct contact system with helium GM-JT

The analysis has been performed for the direct contact system with the helium GM-JT

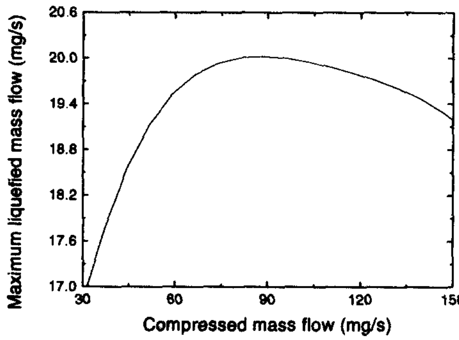

Fig. 8 Maximum liquefaction rate vs mass flow rate in system of Fig. 1(d) with CTI 1020CP when ε =0.93.

Table. 2 Optimal conditions and maximum liquefaction rate for two different cryocoolers and two different HX ε's in system of Fig. 1(c) and (d)

			Direct contact (Fig. 1(c))				Precooled L-H (Fig. 1(d))			
			CTI 1020CP		CTI 1050CP		CTI 1020CP		CTI 1050CP	
HX Effectiveness: ε			0.93	0.99	0.93	0.99	0.93	0.99	0.93	0.99
Optimal conditions	Precooling temperature (K)	T_{PC1}	51.07	69.51	37.24	49.97	131.4	146.8	115.0	139.3
		T_{PC2}	12.41	19.18	13.47	19.39	69.38	96.73	71.47	106.6
	Precooling load (W)	Q_{1ref}	16.08	31.97	12.34	33.0	63.54	69.14	97.97	112.5
		Q_{2ref}	5.45	10.85	3.45	8.35	34.74	41.36	34.14	42.87
	High pressure (bar)		•	•	•	•	110	128	141	123
	Mass flow rate (mg/s)		•	•	•	•	92	278	153	539
Maximum liquefaction rate (mg/s)			5.1*	10.7	3.7	10.3	20.0	26.14	27.22	35.88
Liquid yield: y			•	•	•	•	0.217	0.094	0.178	0.067

* In a good agreement with experimental observation [12,15]

refrigerator, where the two-stage GM cooler is the Models 1020CP and 1050CP of CTI. Figure 10 shows how the liquefaction rate of hydrogen varies according to the mass flow rate of helium through the JT circuit. In the analysis, the high pressure is 20 bar, and the effectiveness of the heat exchangers is in the range between 0.9 and 0.99. As the mass flow rate of helium increases, the liquefaction of hydrogen increases to a certain point and then decreases so that there is a maximum liquefaction.

The maximum liquefaction rate is about 11.18 mg/s in case of 1020CP and 10.93 mg/s in case of 1050CP. The results indicate that no significant improvement is expected by the helium GM-JT refrigeration. The main reason for this is that the helium temperature is not low enough to produce an effective refrigeration by the JT expansion. This configuration will be recommended only if the operating temperature at

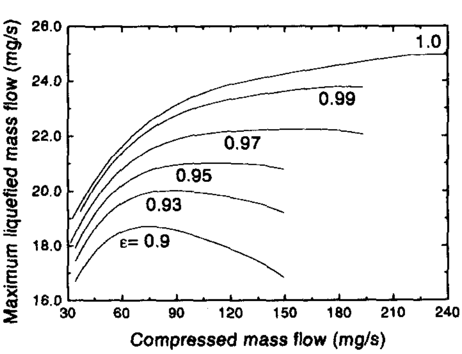

Fig. 9 Maximum liquefaction rate vs mass flow for various ε's, in system of Fig. 1(d) with CTI 1020CP.

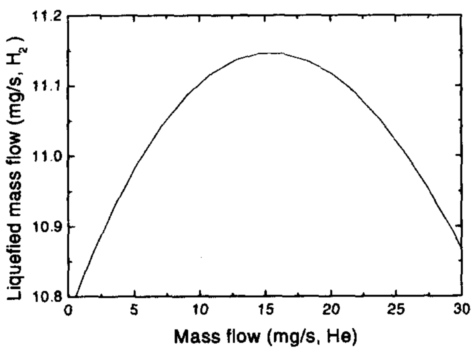

Fig. 10 Liquefaction rate of hydrogen vs mass flow rate of helium in system of Fig. 2 with CTI 1020CP.

the second stage of the GM cooler is below 10K.

4. Conclusions

Several options have been examined to use a Gifford-McMahon cryocooler for the purpose of liquefying hydrogen. For the individual option, a comprehensive thermodynamic analysis has been developed to predict the liquefaction rate by incorporating the typical cooling performance of some commercial GM cryocoolers, the actual thermodynamic properties of hydrogen and the heat exchanger effectiveness.

For the Linde-Hampson liquefaction system precooled by a single-stage or a two-stage GM cooler, it has been demonstrated that there exist unique optimal values for the high pressure and the mass flow rate of compressed hydrogen. The maximum liquefaction rate turns out to be only 4 to 6 times the direct-contact liquefaction with the typical two-stage GM cooler, whose the cold head temperature is below the boiling temperature of hydrogen. The main reason for this poor performance is the limited cooling capacity at the first stage. It can be stated that the Linde-Hampson system precooled by the two-stage GM is practically effective only if the GM cooler has a much greater cooling capacity at around 70-100 K than the currently available models.

Among another options considered in this study, the helium GM-JT refrigerator can be a feasible configuration in the sense that the helium system is operated at pressures much lower than the hydrogen L-H systems. However, the liquefaction performance is not predicted to be enough either, when compared the direct-contact with the two-stage GM. The part of the reason for this is that the precooling temperature of the GM is not low enough for helium to be effective through JT expansion.

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