Development of Cascade Non-Flammable Mixed Refrigerant Joule-Thomson Refrigerator for 100 K

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ABSTRACT

In this paper, a cascade type non-flammable mixed refrigerant (MR) Joule-Thomson (JT) refrigerator is presented to achieve a cooling temperature of 100 K. The working fluid utilized in the main stage (low temperature cycle) of the MR JT refrigerator is composed of nitrogen (N₂), argon (Ar), Tetrafluoromethane (R14) and Octafluoropropane (R218). An azeotropic mixture of R32 and R125 (R410A) is used for the precooling stage (high temperature cycle) refrigerant where the cooling temperature is nearly 240 K. The design specifications of the refrigerator are modeled by using a commercial software with the Peng–Robinson equation of state (EOS). The maximum COP of the optimized MR JT refrigerator is 0.24 at a temperature of 106.6 K. Also, this MR JT refrigerator is fabricated and tested to investigate the refrigeration performance in comparison with the simulated results. The compression system of the refrigeration cycle is implemented by modifying commercial air-conditioning compressors. The R410A precooling stage is simulated by a commercial Stirling cryocooler. The feasibility of the non-flammable MR in the MR JT refrigerator is validated by achieving a minimum temperature of 97.8 K, which is below the freezing point of pure R218 (125 K).

INTRODUCTION

Low temperature cooling technology, namely cryogenic engineering, draws a lot of attention from the technological area such as high temperature superconductor (HTS) applications, liquefied natural gas (LNG) and bio-preservation.

Cryogenic refrigeration technologies for large cooling capacities often adopt the Joule-Thomson (JT) refrigeration cycle. In a JT refrigeration system, a simple JT valve is installed for the expansion process instead of a mechanical expander. Thus, the reliability of JT refrigeration systems are higher than other refrigeration systems due to no moving parts at low temperatures. On the other hand, the low efficiency is an unavoidable drawback of JT refrigeration system because of the irreversible process at the JT expansion part.

Mixed refrigerants (MR) are normally applied in the JT refrigerators to boost their efficiency. Generally, MR are composed by the combination of low-boiling point refrigerants and high-boiling point refrigerants. Low-boiling point components are effective to achieve a lower cooling temperature whereas the high-boiling point components accomplish boosting the refrigeration efficiency.

Thus, the MR should be optimized with the target heat absorption temperature before constructing the MR JT refrigeration cycle. Since the flammable refrigerants have high JT coefficients and isothermal enthalpy differences compared to those of the non-flammable refrigerants, the flammable MR are favorable for utilization in MR JT refrigerators. Therefore, the previous studies commonly facilitated using flammable MR such as methane, ethane, propane and butane [1]. Nowadays, MR JT refrigerators are frequently applied in the natural gas liquefaction plants where the lowest temperature was approximately 110 K [2, 3]. On the other hand, flammable MR have critical drawbacks which are the flammability and the explosiveness. Non-flammable MR are attractive in this sense and start to be applied in MR JT refrigerators.

Unfortunately, there are only a few studies about non-flammable MR JT refrigerators. Boiarski et al., investigated the performance of a small-scale MR JT refrigerator by using both flammable MR and non-flammable MR in 2005 [4]. They mentioned that the freezing temperature of the non-flammable MR tested is higher than 110 K. Khatri and Boiarski discussed the development of a non-flammable single stage MR JT refrigerator where the target temperature was below 110 K, in 2008 [5]. They also mentioned that the cooling temperature of 110 K is limited by the high boiling components related to freezing temperature, and poor thermodynamic efficiency is achieved compared to using flammable MR. Thus, non-flammable MR presented a low thermodynamic efficiency to reach 100 K or below.

This paper focuses on the development of a high efficiency non-flammable MR JT refrigerator to achieve a cooling temperature of 100 K. The freezing limitation of the working fluid should be identified and therefore, the freezing point for non-flammable MR was studied in a previous study [6]. The design result for achieving the maximum Coefficient of Performance (COP) was also described in the previous studies [7]. This paper discusses the experimental verification of the design result, and the detailed description of a capillary tube as the expansion part.

DESIGN RESULT OF CASCADE MR JT REFRIGERATOR

The design of a non-flammable MR JT refrigerator is divided into two parts. First, the freezing limitation of the target non-flammable MR should be identified. Freezing points of the target non-flammable MR to achieve the 100 K cooling temperature was verified in a previous study [6]. The selected components are Nitrogen (N₂), Argon (Ar), Tetrafluoromethane (CF₄, R14) and Octafluoropropane (C₃F₈, R218). The freezing points of the mixture of Ar, R14 and R218 are experimentally verified according to the various molar compositions. N₂ is excluded in the experiments because its normal boiling temperature is lower than the target cooling temperature. Certain ternary MR (Ar, R14 and R218) have a measured freezing temperature below 77 K [6]. This freezing temperature is much lower than the triple point temperature of R218 (125 K). The constraint of the molar composition for quarternary MR (N₂, Ar, R14 and R218) is established according to a previous study.

Second, the optimal operating condition of a non-flammable MR JT refrigerator is designed by commercial software (Aspen HYSYS 8.0) with the aforementioned composition constraint [7]. The calculation was conducted for three different configurations: (1) the single stage with ternary MR (Ar, R14 and R218), (2) the single stage with quaternary MR (N₂, Ar, R14 and R218), and (3) the 2-stage (cascade type) with quaternary MR. Figure 1 (a) shows the schematic diagram of the single stage configuration and Figure 1 (b) displays the cascade type configuration [7]. Also Figure 1 (b) depicts the calculation points or instrumentation positions for this configuration.

The target cooling temperature is set at approximately 100 K, and the variables are selected as the mass flow rate of the working fluid, the discharge pressure, the suction pressure, and the molar composition. A cascade type configuration with a quaternary MR presents the highest COP of the entire calculation result. The molar composition of quaternary MR was N2:Ar:R14:R218 = 0.2:0.3:0.3:0.2 in the simulation with the maximum COP. Table 1 shows the design result of the cascade type non-flammable MR JT refrigerator with the highest efficiency. Each position of Table 1 is illustrated in Figure 1 (b), as mentioned before. The simulation assumes a two-stage compression and its isentropic efficiency is 80%. The pressure drop inside the heat exchanger is neglected.

The lowest temperature of this cascade MR JT refrigerator is calculated to be 96.3 K and the temperature after the evaporator is 106.6 K. The normalized cooling capacity is attained as 19.5 J/g,

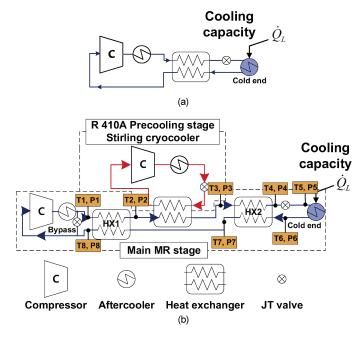


Figure 1. Schematics of (a) single stage configuration and (b) cascade type MR JT refrigerator configuration with calculation point or measurement position.

which is the cooling capacity divided by the mass flow rate of the working fluid. The normalized compression work is also obtained as 73.7 J/g, and the normalized precooling work (precooling work / mass flow rate of main MR) was calculated as 6.7 J/g. The maximum COP of this cascade type non-flammable MR JT refrigerator is calculated to be 0.24. One of the important points is that the thermodynamic state of position 4 is a subcooled liquid state. This will be discussed later in this paper.

Table 1. Design results of cascade type non-flammable MR JT refrigerator for 100 K cooling temperature

Position	Temperature [K]	Pressure [kPa]	Vapor quality (kg/kg)
1	300	1400.0	Superheated
2	254.2	1400.0	0.973
3	247.0	1400.0	0.912
4	111.1	1400.0	Subcooled
5	96.3	220.0	0.046
6	106.6	220.0	0.314
7	240.0	220.0	Superheated
8	297.0	220.0	Superheated
Condition		Value	Comment
Molar composition [-]		N ₂ :Ar:R14:R218 = 0.2:0.3:0.3:0.2	
Cooling capacity [J/g] (A)		19.5	
Compression work [J/g] (B)		73.7	
Precooling work [J/g] (C)		6.7	Cooling Temperature : 244 K Refrigerant: R410A
COP [-]		0.24	A/(B+C)

EXPERIMENTAL APPARATUS OF CASCADE MR JT REFRIGERATOR

The experimental apparatus is composed of two parts, the compressor system and the cold box.

Compression system

Figure 2 (a) shows the complete view of the developed compressor system, and Figure 2 (b) presents the assembly of each component in the compressor system. The compression system is developed with two commercial air-conditioning rotary vane compressor units (GPD420DAA and GPD240DAA, LG electronics) and an oil removal system [8]. The oil removal system utilizes two oil separators, a water-chilled aftercooler, oil mist filters, and a filter drier. A four-stage compressor system is constructed to achieve a sufficiently high compression ratio (the ratio between the discharge pressure and the suction pressure). The swept volume of the first compressor (GPD420DAA) is 40.5 cc/rev and that of the second compressor (GPD240DAA) is 23.5 cc/rev. Since the different volume flow rates of the two compressors require different operating frequencies, commercial inverters (Yaskawa cooperation, model A1000) control the operating frequencies of the two compressor units. An oil removal system is composed of oil separators (Emerson AW), oil mist filters and a filter drier, to prevent the oil and moisture in the working fluid.

Cold box

The cold box includes several heat exchangers, the evaporator, the capillary tube and the cryo-cooler instead of the precooling stage. The entire cold box system is attached to the top flange of the vacuum chamber. Figure 3 (a) depicts the developed cold box system and Figure 3 (b) illustrates the cold box system after the multi-layer insulation (MLI) installation.

Heat exchanger 1 (HX1) and heat exchanger 2 (HX2) are the diffusion bonded printed circuit heat exchangers (PCHEs). Stainless steel plates are applied to construct this PCHEs. The single channel is 300 µm in height and 300 µm in width. Each layer has 10 channels and is in a meandering shape. The channel layer has a height of 210 mm and a width of 77 mm. A 20-layer (10 layers for hot side and 10 layers for cold side) PCHE is fabricated for HX 1, and a 30-layer (15 layers for hot side and 15 layers for cold side) PCHE is used for HX 2. The entire layers are fabricated by using a diffusion bonding process.

A Stirling cryocooler (Sunpower Inc, Cryotel CUBE) accomplishes precooling of the MR JT system in the experiment. A machined copper block surrounded with a copper tube is attached for heat exchange. The working fluid of main MR cycle is precooled in this copper tube. The precooling temperature (position 3) is controlled by the cryocooler and the auxiliary heater at the cold tip. The temperature at position 3 is maintained between 240 K and 248 K.

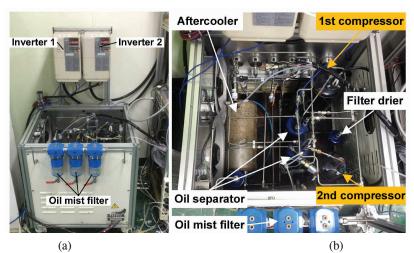


Figure 2. Photographs of compression system (a) the complete view and (b) the detailed compression system.

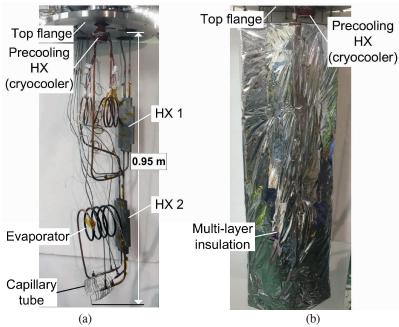


Figure 3. Photographs of experimental apparatus for non-flammable cascade MR JT refrigerator; (a) the detailed view and (b) the covered view with MLI

The JT expansion part is constructed with a capillary tube to avoid a clogging problem. The capillary tube is made of a stainless steel tube, which is 1.6 mm (1/16 inch) in the outer diameter and 1.2 mm in the inner diameter. The selected lengths of capillary tubes are 1.5 m and 3.0 m. The evaporator is fabricated with a coiled 6.4 mm (1/4 inch) copper tube and the heater is covered with Stycast 2850FT epoxy.

The whole cold box system is insulated by a vacuum chamber and multi-layer insulation to prevent heat flow from the environment. Ten layers of MLI (RUAG GmbH, Coolcat 2NW) are applied to cover the entire cold box system.

In the cold box system, eight temperature sensors and eight pressure transducers were installed to measure the temperature and pressure of each location as seen in Figure 1 (b). Silicon diode type temperature sensors (Lakeshore Inc, DT-670-SD) are used, and FP 2000 pressure transducers (Honeywell Inc) are selected. To measure the mass flow rate of the working fluid, a Coriolis mass flow meter (Bronkhorst, CORI-FLOW M55) is utilized at the discharge of the compressor system. The data is collected by a DAQ system (National Instruments, SCXI-1000, SCXI-1125, SCXI-1320 and SCXI-1328) and a desktop computer. The measurement position is illustrated in Figure 1(b), as mentioned earlier.

EXPERIMENTAL RESULT AND DISCUSSION

1.5 m capillary tube experimental result

The 1.5 m long capillary tube is first applied to the experimental apparatus. Figure 4 shows the typical cool down characteristics of the non-flammable MR JT refrigerator. The composition of working fluid is the same as the design result (N₂:Ar:R14:R218 = 0.2:0.3:0.3:0.2). Figure 4 (a) shows the temperature data of the experimental results and Figure 4 (b) presents the pressure and mass flow rate data of the results. Figure 4 (c) shows the effectiveness of each heat exchanger based upon the temperature data. The lowest temperature obtained is 103 K, which is higher than the design result. The considerably higher cooling temperature is due to the relatively high pressure drop at the low pressure stream. The unexpected large pressure drop causes a pressure increase at position 5, which is higher than the design result.

Figure 4 (d) represents the quality profile of all experimental results. The quality of the working fluid is obtained by REFPROP 9.1 software [9]. For the sake of convenience, the pseudo-quality value is assumed as 1.05 and -0.05 for the superheated state and the subcooled state, respectively. The working fluid mass flow rate is drastically increased when the state at position 4 (inlet of the capillary tube) reaches a subcooled liquid state (approximately 7 hours later). The pressure at position 4 is decreased considerably when the temperature at position 4 gradually reduces.

The reason for the pressure reduction at position 4 can be explained by two effects, which are the complete liquefaction of the working fluid and the lack of pressure drop in the capillary tube. The working fluid density is greatly increased when the MR is fully liquefied. Therefore, the liquid pressure can be reduced. Furthermore, the pressure drop in the capillary tube was weaken because the state of the working fluid transforms from a two-phase state to a liquid state, at the inlet of the expansion part. The pressure drop at the capillary tube should be increased to maintain the stable operation. Thus, a 3.0 m long capillary tube is added at the expansion part in the same experimental apparatus and tested in the same operating conditions.

3.0 m capillary tube experimental result

Figure 5 displays the experimental results with the 3.0 m capillary tube expansion part. The lowest temperature in this case is maintained as 97.8 K, which is 5.2 K lower than that of the system with the 1.5 m capillary tube. When the state at position 4 enters a subcooled state, the mass flow rate significantly increases, as mentioned in the aforementioned test result. The mass flow rate is slightly reduced at the steady state therefore, the pressure drop at the low pressure stream is

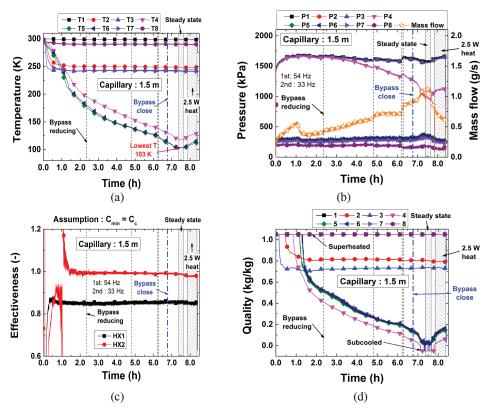


Figure 4. Cool down characteristics of the cascade type non-flammable MR JT refrigerator (N2:Ar:R14:R218 = 0.2:0.3:0.3:0.2) using the capillary tube length of 1.5 m; (a) temperature profile, (b) pressure and mass flow rate profile, (c) heat exchanger effectiveness profile and (d) quality profile at each location.

slightly reduced. Thus, the pressure at position 5 (i.e. pressure after the capillary tube) is maintained at approximately 230 kPa. Although the lowest temperature is reached as designed, the pressure at position 4 is too low, which shows a similarity to shorter length of capillary tube.

Characteristics of capillary tube

In this study, two different capillary tube lengths are reported with the same experimental apparatus. One is 1.5 m and the other is 3.0 m, each has the same diameter and cross-sectional area. The pressure drop of each capillary tube will be different at the non-choked condition. Nevertheless, the maximum mass flow rate for each result is close to 1 g/s and the pressure difference at the capillary tube is approximately 700 kPa. Figure 6 (a) and (b) plot the mass flow rate and the quality measurement of each experiment and Figure 6 (c) and (d) show the pressure profile between the inlet (P4) and the outlet of the capillary tube (P5). Although the lengths of capillary tubes are different for the two experiments, the similar pressure and mass flow rate profiles are obtained from the experiment. These results indicate that if the inlet condition reaches the subcooled liquid state, the choking phenomenon would occur in the capillary tube. While the temperature of position 4 gradually drops, the pressure and mass flow rate are gradually decreased after the choking condition. This characteristic is due to the fact that the density is increased according to the enhanced subcooling. The escalation of the mass flow rate at the choking condition leads to the pressure cutback at the position 4, and this reduced pressure causes the mass flow rate reduction in the next step. Thus, a higher degree of subcooling and controllability of the mass flow rates (i.e. by using valves) is required for achieving a stable operation of the subcooled JT expansion.

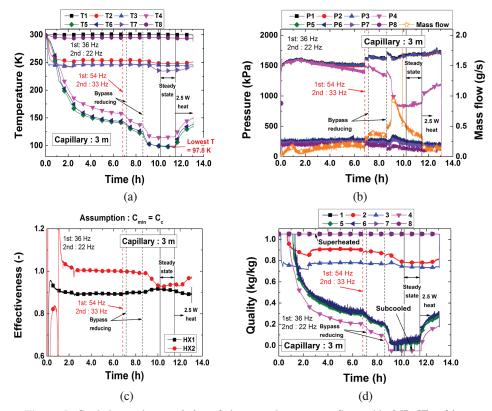


Figure 5. Cool down characteristics of the cascade type non-flammable MR JT refrigerator (N2:Ar:R14:R218 = 0.2:0.30.3:0.2) using the capillary tube length of 3.0 m; (a) temperature profile, (b) pressure and mass flow rate profile, (c) heat exchanger effectiveness profile and (d) quality profile at each location.

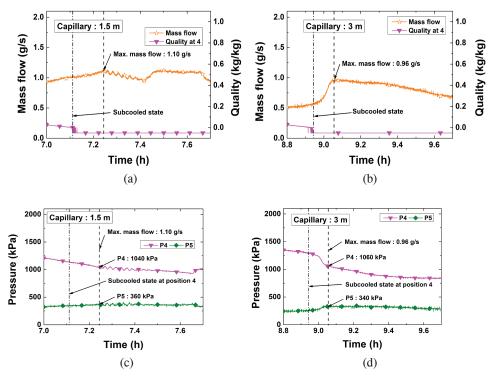


Figure 6. Mass flow rate, quality and pressure profile at the start of subcooled state before the expansion part; (a) mass flow rate and quality profile of 1.5 m capillary tube, (b) mass flow rate and quality profile of 3.0 m capillary tube, (c) pressure profile between the inlet and the outlet of the 1.5 m capillary tube and (d) pressure profile between the inlet and the outlet of the 3.0 m capillary tube (N2:Ar:R14:R218 = 0.2:0.3:0.3:0.2).

Cold end characteristics

By the design result, the target cooling capacity was calculated as 19.5 J/g at 106.6 K. Before the full loading of the design result (approximately 10 W), 2.5 W heat is applied after the steady state for each experiment. Figure 7 (a) and (b) display the temperature profiles and the quality profile of the 3.0 m capillary tube, respectively. Figure 7 (c) displays the P4 (capillary tube inlet), saturated liquid pressure calculated by T4, and the mass flow rate of the experimental result [9]. As can be seen that the MR JT refrigerator cannot endure the poor cold duty. When the tiny heat is applied in the evaporator, the mass flow rate of the working fluid is drastically reduced. This reason of the mass flow rate reduction is that the tiny heat flow changes the state at position 4, which has a tiny degree of subcooling. When the heat is applied at the evaporator, the state of the capillary inlet (location 4) becomes two-phase instead of subcooled because T4 is increased due to the increased value of T6. Then the pressure drop inside of the capillary tube is drastically raised, and the mass flow rate is decreased to maintain the certain pressure drop. As a chain reaction, the mass flow rate reduction lowers the cooling capacity at the cold end, and the lowest temperature of the cold end drastically increases. Thus, the degree of subcooling at the inlet of JT expansion part must be sufficiently developed to construct the subcooled JT expansion and the stable operation of the MR JT refrigerator.

CONCLUSION

In this paper, a cascade type non-flammable MR JT refrigerator is developed and tested according to the design result. Two types of capillary tubes are applied to construct the JT expansion part. The lowest temperature of the developed MR JT refrigerator is 97.8 K, and some interesting

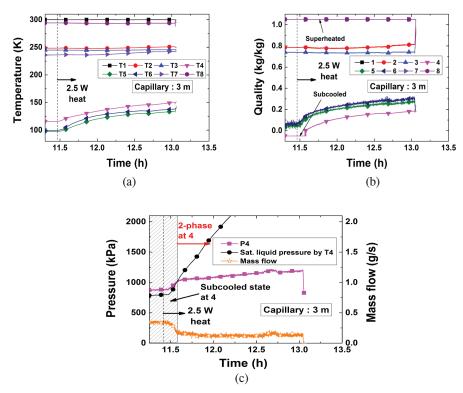


Figure 7. Temperature and quality profiles after heating for 3.0 m capillary tube; (a) temperature profile ,(b) quality profile of 3.0 m capillary tube, (c) saturated liquid pressure, capillary inlet pressure (P4) and mass flow rate profile (N2:Ar:R14:R218 = 0.2:0.3:0.3:0.2)

characteristics and limitations have been observed in association with using a capillary tube during the refrigerator operation.

- ➤ The selected non-flammable mixed refrigerant of N₂, Ar, R14 and R218 does not freeze at 97.8 K, which is much lower than the freezing temperature of R218 (125 K).
- ➤ Choking phenomenon occurs in the capillary tube. The length of capillary tube does not affect the mass flow rate of the working fluid.
- > The degree of subcooling must be sufficiently secured for stable operation when the subcooled liquid JT expansion is required at the JT expansion part.
- ➤ To achieve higher degree of subcooling and controllability of the mass flow rate, a valvecontrol is necessary for this kind of MR JT refrigerator.

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REFERENCES

Chakravarthy V. S., Shah R. K., Venkatarathnam G. "A Review of Refrigeration Methods in the Temperature Range 4–300 K," *Journal of Thermal Science and Engineering Applications*, 3, no. 2 (2011), pp. 020801.

- 2. Venkatarathnam, G., Cryogenic Mixed Refrigerant Processes, Springer, New York (2008), pp.145-153.
- 3. Lim, W., Choi, K., Moon, I., "Current Status and Perspectives of Liquefied Natural Gas (LNG) Plant Design". *Industrial & Engineering Chemistry Research*, 52, no. 9 (2012), pp. 3065-3088.
- 4. Boiarski, M., Podtcherniaev, O., Flynn, K., "Comparative Performance of Throttle Cycle Cryotiger Coolers Operating with Different Mixed Refrigerants in the 13th International Cryocooler Conference," *Cryocoolers* 13, Springer US, USA (2005), pp. 481-488.
- 5. Khatri, A., Boiarski, M., "Development of JT coolers operating at cryogenic temperatures with non-flammable mixed refrigerants in the Cryogenic Engineering Conference," (2008). pp. 3-10.
- 6. Lee, C., Yoo, J., Lee, J., Jeong, S., "Visualization of the solid–liquid equilibria for non-flammable mixed refrigerants". *Cryogenics*, 75, no. 0 (2016), pp. 26-34.
- Lee, C., Jin, L., Jeong, S., "Design of non-flammable mixed refrigerant Joule-Thomson refrigerator for precooling stage of high temperature superconducting power cable in the 8th Asian Conference of Applied Superconductivity and Cryogenics," *Applied Superconductivity - Proceedings of ACASC 2015*, China (2015).
- 8. Lee, J., Oh, H., Jeong, S., "Investigation of neon–nitrogen mixed refrigerant Joule–Thomson cryocooler operating below 70 K with precooling at 100 K," *Cryogenics*. 61, no. 0 (2014), pp. 55-62.
- Lemmon, E.W., Huber, M.L., M.O.McLinden. "NIST Standard Reference Database 23: Reference fluid thermodynamic and transport properties-REFPROP 9.1," National Institute of Standards and Technology, Standard Reference Data Program; 2013.