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Mixed refrigerants for a glass capillary micro cryogenic cooler ☆

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ABSTRACT

Optimized mixed refrigerants are applied in Joule–Thomson (JT) micro cryogenic coolers (MCC) to enhance efficiency. Mixed refrigerants deliver equivalent refrigeration power with much lower pressure ratio and flow rate compared to pure nitrogen refrigerant. To determine the behavior of mixtures in MCCs, the normal boiling points of the components, mixture solubility, and refrigeration loss due to pressure drop on the low-pressure side of the heat exchanger are evaluated. The MCC discussed here was designed to operate at 77 K with the heat exchanger warm end precooled to 240 K by a thermo-electric cooler. An optimized five-component mixed refrigerant was calculated to provide a minimum isothermal enthalpy difference of 1.35 kJ/mol between 77 K and 240 K with a high pressure of 1.6 MPa and a low pressure of 0.1 MPa. Experimentally, a stable temperature of 140 K was achieved with a flow rate of 11 μmol/s. A transient temperature of 76 K was observed.

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Micro cryogenic coolers have drawn considerable attention from researchers and application developers due to their small volume, low noise, fast response, and potential to be portable and mobile. Prior research [1–5] on MCCs used pure gas refrigerants (single-component). However, pure gas refrigerants require high pressure ratios and high flow rates, which require large compressors. This increases the size of the whole system and limits mobility for MCC applications. In this work, an optimized five-component mixed refrigerant is applied in Joule–Thomson (JT) micro cryogenic coolers (MCC). It is shown that compared to pure nitrogen, the mixed refrigerants can deliver equivalent refrigeration power with much lower pressure ratio and flow rate. The benefit of the mixed refrigerant solution provides potential opportunities for MCCs to integrate with miniature or micro compressors. For example, commercially available miniature compressors [6] can be serially linked to provide up to 20:1 pressure ratio. This mobility can be greatly beneficial to applications such as cooling IR sensors, low noise amplifiers, MEMS resonators, RF front ends, magnetic sensors, chemical sensors, high Tc superconducting devices and innovations people have not yet begun to explore.

In the 1980's, Little et al. [1–3] first successfully demonstrated JT MCCs fabricated by bonded etched glass wafers. With a high pressure of 16.5 MPa and 107 μmol/s nitrogen gas (3 mg/s), it achieved 88 K in 30 s. A high pressure gas tank was used to supply nitrogen for the cooler. Recently, Lerou et al. [5] cooled MCCs by using 8.0 MPa nitrogen input and expanding to 0.6 MPa. The cold head reached 100 K with 12 mW net refrigeration power. During development of MCCs, Lerou et al. [5] encountered serious clogging problems in their micro heat exchanger or micro JT valves caused by freezing water vapor. Pure ethylene at a high pressure of 2.0 MPa was also used as a refrigerant in a MCC by Burger et al. [4]. With 238 K thermoelectric precooling, the cooler reached 170 K with 200 mW net refrigeration power and 17.8 μmol/s (0.5 mg/s) flow. Both Lerou and Burger's coolers are to be connected to a fairly large sorption compressor for high pressure gas supply.

In macro Joule–Thomson refrigeration systems, mixed refrigerants are widely applied to enhance the efficiency and boost refrigeration power. Missimer [7], Radebaugh [8], and Boiarski [9] have reviewed recent developments and history of mixed refrigerants. Fuderer and Andrija [10] first used mixed gases in a single stream without phase separators in 1969. They found that the mixtures experienced mostly two-phase flow in the heat exchanger. As a result, boiling and condensing heat transfer of two-phase flow greatly enhances cooling efficiency. Boiarski and Longthworth [9] have demonstrated mixtures for 67 K cooling for a JT system with only 2 MPa pressure applied. They also pointed out that a possible liquid–liquid separation of the nitrogen from hydrocarbons in the mixtures happens at extremely low temperature if the pressure is lower than 6 MPa. To quantify its performance, Marquardt et al. [11] further developed models and optimization approaches for mixtures. Little [12,13] also verified the great enhancement in refrigeration power using mixtures compared with pure nitrogen.

JT systems rely on the fact that there is a reduction of enthalpy with increased pressure in non-ideal gases. The gross refrigeration power $\dot{q}$ is given by [8]:

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$$\dot{q} = \dot{n}(\Delta h_T)_{\min} \quad (1)$$

where $\dot{n}$ is the molar flow rate and $(\Delta h_T)_{\min}$ is the minimum isothermal enthalpy difference between the high pressure and low pressure enthalpies within the temperature range of interest. In fluids, the largest enthalpy difference usually occurs at or close to the temperature of the phase change from liquid to gas. For pure refrigerants, $(\Delta h_T)_{\min}$ occurs at the highest temperature of interest which is the warm end of the heat exchanger. As a result, several cooling stages and different gases have to be applied to enhance efficiency to reach cryogenic temperatures. In mixed refrigerants, components are selected with boiling points across the temperature range of interest. By controlling the amount of different components in a mixture, the enthalpy difference is made more uniform across the temperature range, and $(\Delta h_T)_{\min}$ is maximized. Table 1 lists $(\Delta h_T)_{\min}$ for pure nitrogen and mixtures under different pressure ratios. The five-component mixed refrigerant has the largest $(\Delta h_T)_{\min} \sim 1.35$ kJ/mol. To deliver 15 mW of gross refrigeration power, JT cryocoolers using mixed refrigerants only require 1.6 MPa pressure input and 11 μmol/s or 15 sccm flow, while those using pure nitrogen require about three times the pressure input and flow as shown in the first row in Table 1.

Fig. 1 shows the schematic of a glass capillary JT micro cryocooler and photos of a glass capillary MCC. The schematic illustrates the cold head with a JT expansion valve, the counter flow heat exchanger (CFHX), and the precooler. The coaxial CFHX is assembled by using hollow-core glass fibers (ID/OD = 75 μm/125 μm) as inner fluid channels and enclosed by a glass capillary tube (ID/OD = 536 μm/617 μm). As shown in the photo, the CFHX is 25 mm in length, and the cold head is 2 mm square. To operate the cooler, the high pressure mixed refrigerant at 1.6 MPa is precooled to 240 K and sent through the glass fibers to the cold head. In the JT expansion orifice, the refrigerant experiences an isoenthalpic expansion causing it to cool. The expanded low pressure and cooled refrigerant then returns back through the interstitial spaces between the fibers in the capillary tube to precool the incoming warm high pressure mixture to enhance efficiency. The whole system is vacuum packaged with a radiation shield to minimize conduction and radiation heat transfer from the surrounding. A compressor is used to complete the JT cycle into a closed loop.

Fig. 2 shows the plot of the isothermal enthalpy difference of an optimized five-component mixed refrigerant. The optimized mixed

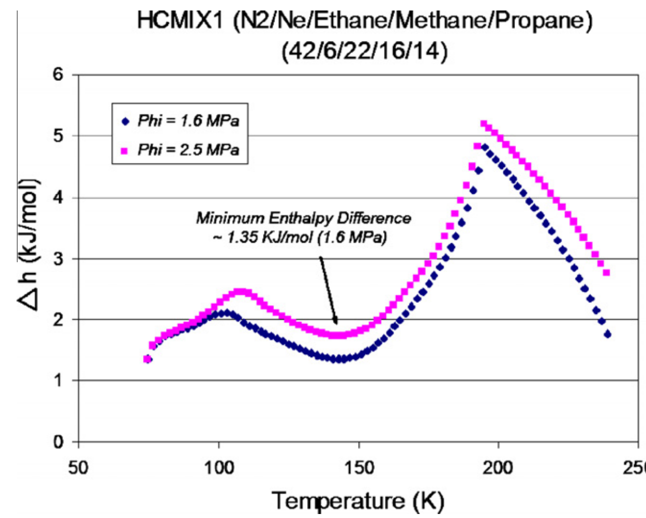

Fig. 2. Enthalpy difference vs. temperature of an optimized five-component mixed refrigerant. Both enthalpy difference of 0.1 MPa (1 bar)–1.6 MPa (16 bar) and 0.1 MPa (1 bar)–2.5 MPa (25 bar) are plotted as a function of temperature.

Table 1
Minimum enthalpy difference of pure nitrogen and mixed refrigerants over the temperature range of 77 K to 240 K for the high pressure P_H and low pressure P_L are shown in the table. Also shown are the ideal COP, the efficiency, and required flow rate for each refrigerant to provide 15 mW of gross refrigeration power from Eq. (1).

Refrigerant	P_H (MPa)	P_L (MPa)	$(\Delta h_T)_{\min}$ (kJ/mol)	COP_{ideal}	η (%Carnot)	$\dot{n}$ (μmol/s)
N_2	5.0	0.1	0.468	0.0480	14.4	32.0
N_2	2.5	0.1	0.232	0.0289	8.7	64.6
N_2	1.6	0.1	0.146	0.0211	6.3	102.7
5-comp mix	1.6	0.1	1.35	0.2497	52.9	11.1

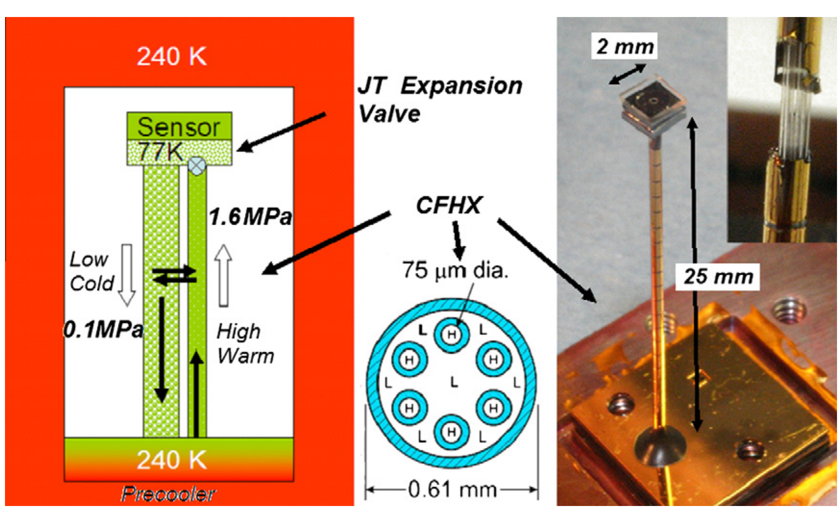

Fig. 1. Schematics and photos of the MCC. The left schematic illustrates the MCC key components, which are the JT expansion valve, the CFHX, and the precooler. High pressure warm gas is pumped through the fibers (as shown in cross sectional views) in the CFHX to the JT expansion valve. The expanded cooled gas returns back in the interstitial spaces in the CFHX to precool the incoming gas. The direction of flow is indicated with arrows. Photos of the MCC assembly and fibers in the capillary are shown on the right hand side.

refrigerant is designed using NIST software known as NIST4 [14]. The mixture in terms of mole fraction consists of 14% propane, 16% ethane, 22% methane, 42% nitrogen, and 6% neon, and their normal boiling points are 231 K, 184.6 K, 111.6 K, 77 K, and 24 K, respectively. The minimum enthalpy difference between 0.1 MPa and 1.6 MPa (~1.35 kJ/mole) occurs at 140 K.

The solubility of each component has to be evaluated to ensure that no component freezes during cooling or liquefaction. Fig. 3 shows the relationship between normal boiling point and ideal solid solubility in nitrogen at 75 K of commonly used components in mixtures. Based on this simple idealized calculation, the solid solubility at 75 K of methane, ethane, and propane are 76%, 45%, and 50% respectively. As a result, the light hydrocarbon components in our mixture should be soluble in nitrogen at temperatures higher than 75 K.

Pressure drop in heat exchangers can cause refrigeration loss in cryocooler systems. The minimum enthalpy difference and hence refrigeration power can be decreased due to pressure loss in micro heat exchangers. Significant pressure drop can occur on the low pressure returning channel. Little [15] reported encountering problems of refrigeration loss due to low-side pressure drop during their early development of miniature JT micro-cryocoolers. They solved it by making the returning flow laminar through re-designing micro channels. Fig. 4 shows the relationship between the minimum enthalpy difference and the low-side pressure of the mixed refrigerant. The mixture has higher minimum enthalpy difference when the low-side pressure is lower. Two regions can be identified in the plot. First, the minimum enthalpy difference increased only slightly when the pressure decreases below 0.1 MPa. Second, the minimum enthalpy difference drops rapidly when the pressure increases above 0.1 MPa. As a result, vacuum on the low-side has limited influence in the refrigeration power, but a 10% pressure increase on low-side can cause a 13.3% refrigeration power loss.

To test mixed refrigerants in the cold head of a MCC, a macro scale compressor was used to provide the desired low and high pressures across a back-pressure regulator in a bypass loop. The high pressure flow through the MCC cold head was in parallel to the bypass loop. Fluid was precooled to 240 K prior to entering the cold head by a Gifford McMahon cryocooler. The MCC is shielded with 240 K shielding and placed in a vacuum chamber to reduce heat loads due to conduction and radiation from the environment. The high side pressure, low-side pressure, and flow

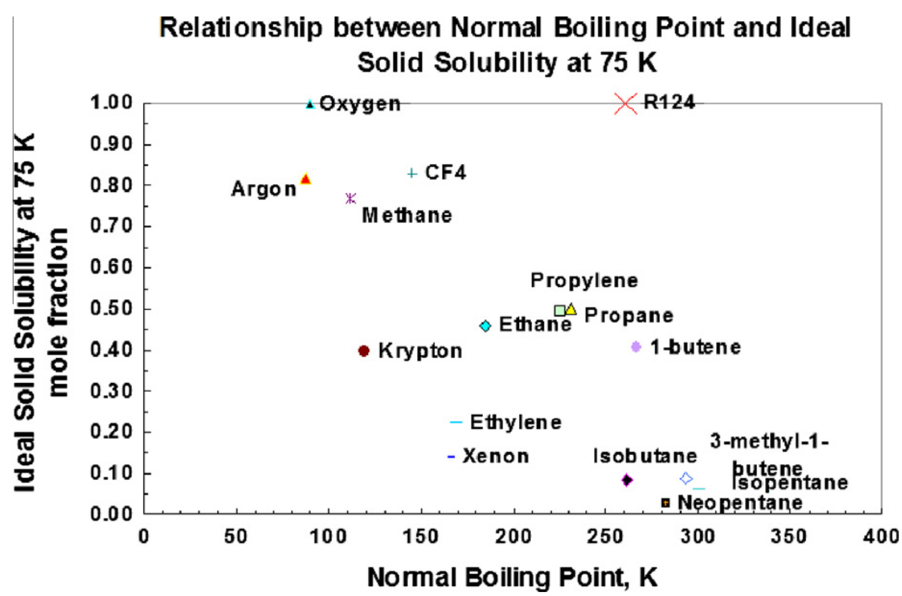

Fig. 3. Ideal solid solubility at 75 K of commonly used components in mixtures as a function of their normal boiling points.

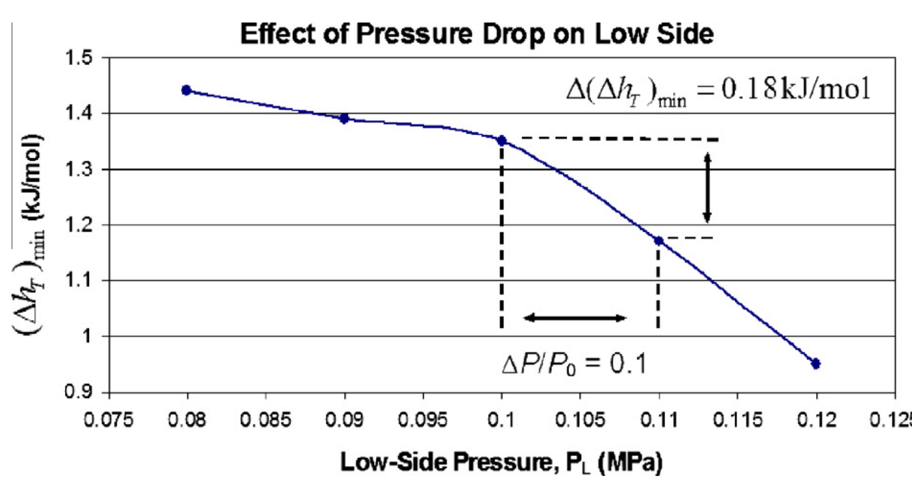

Fig. 4. Effect of pressure drop in the low-side on the minimum enthalpy difference. As shown, a 10% pressure increase on the low-side due to a pressure drop causes 0.18 kJ/mol loss in enthalpy difference.

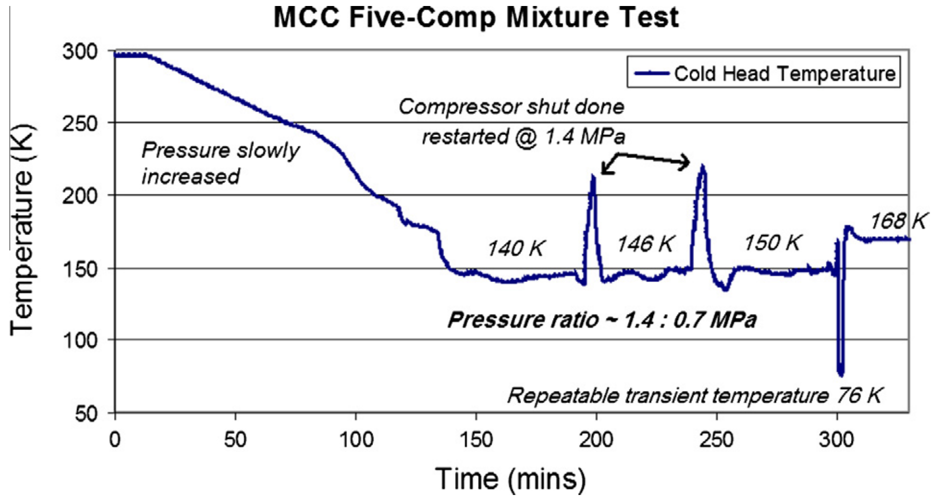

Fig. 5. Cooling test of the MCC using five-component mixed refrigerant. The mixture is precooled to 240 K before sending into the MCC, and the cold head temperature is measured by a thin film Ti thermometer affixed on it.

rate are monitored. The cold head temperature is measured by a thin film Ti resistance thermometer affixed on the tip. The sensor is calibrated against a commercial silicon diode thermometer over the temperature range of 76–300 K. Fig. 5 shows MCC cooling results using the five-component mixed refrigerant. The cold head temperature slowly decreases as the pressure increases. It reached a stable temperature of 140 K with 1.4:0.07 MPa pressure ratio and ~11 μmol/s (15 sccm) flow. By utilizing Eq. (1), a gross refrigeration power of 15 mW is estimated if a pressure ratio of 1.6:0.1 MPa is achieved. However, we are unable to achieve 1.6 MPa in the test. To achieve 77 K stable cooling, the MCC has to be further optimized in the JT expansion valve, the micro heat exchanger, and the thermal isolation design to reduce heat loss and ensure the right pressure ratio and flow rate.

During the test, two unexpected compressor shut downs resulted in two sharp peaks in the figure. Restarting the compressor with 1.4 MPa input showed rapid cooling from 220 to 140 K in less than 5 min. A transient temperature of 76 K was observed after the low-side pressure was rapidly increased by manipulating the pressure regulator.

The 76 K rapidly transient cooling could be caused by an enhanced condensation and local pressure drop on the low pressure returning channel in the micro heat exchanger [16]. When manipulating the pressure regulator to increase the low-side pressure in the heat exchanger, a back-stream room temperature mixture flows into the low-pressure side of the heat exchanger. These warm gas mixtures condense into liquids in the low-side of heat exchanger due to the low temperature and cause a sudden pressure drop locally. The local pressure drop on the low-pressure side of the heat exchanger increases the pressure difference and flow rate across the expansion value. An increase of refrigeration power is occurred and temperature on the cold head dropped rapidly. In Fig. 5, the chart shows a temperature rise prior to the 76 K rapidly cooling due to the warm mixture flowing into the heat exchanger. However, no pressure drop is observed in our test setup since the pressure sensor is comparatively far from the MCC and can hardly detect the local pressure difference in the micro cryogenic cooler. The local pressure drop quickly recovered after flow stabilized. We are currently designing an experimental apparatus to further study mixture behavior in micro channels.

In summary, we demonstrated that optimized mixed refrigerants can be utilized in micro-cryocoolers to enhance efficiency and boost refrigeration power. The normal boiling point of components, mixture solubility, and refrigeration loss due to pressure drop on the low-pressure side are evaluated. With 240 K precooling and 11 μmol/s (15 sccm) flow rate, a stable temperature of 140 K was reached with a 1.4:0.07 MPa pressure ratio using an optimized five-component mixture. A transient temperature of 76 K was repeatably observed due to the enhanced condensation in the micro heat exchanger after the low-side pressure was rapidly increased by manipulating the pressure regulator. Future studies of mixture behavior in micro channels at various temperatures are important to further improve mixture performance and to benefit microcryocooler development.

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