TEMPERATURE INSTABILITY COMPARISON OF MICRO- AND MESO-SCALE JOULE-THOMSON CRYOCOOLERS EMPLOYING MIXED REFRIGERANTS

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ABSTRACT

Previously we demonstrated cryogenic cooling in a Joule-Thomson (JT) microcryocooler (MCC) with mixed refrigerants operating at pressure ratios of 16:1 that achieved stable temperatures of 140 K, with transient temperatures down to 76 K, with precooling of the refrigerant to 240 K. Pre-cooling improves the minimum enthalpy difference, (Δh_T)min compared with that of pure fluids. Micro-scale compressors have been unavailable to meet 16:1 ratios. By reducing the ratio to 4:1, mini-compressors become viable in the near term. Utilizing mixed refrigerants optimized for 4:1 pressure ratios we compare the performance stability of this micro-JT employing a 25 mm long multichannel glass fiber heat exchanger (outer low-pressure capillary ID/OD=536 μm/617 μm, inner high-pressure channels ID/OD=75 μm/125 μm) with a scaled up (meso-scopic) version employing a 20 cm long single channel stainless steel heat exchanger (outer low pressure channel ID/OD=580 μm/760 μm, inner high pressure channel ID/OD=150 μm/266 μm). This easy to fabricate and modify meso-scale version was fabricated to investigate the temperature instabilities of mixed refrigerants for similar operating conditions but for proportionally higher flows of ~ 30 cm^3/min compared with ~ 10 cm^3/min. We compare measured pressures, flow rates, temperatures, and stabilities for both micro- and meso-JT cryocoolers to better understand the causes for the temperature instabilities within the micro-JT cryocooler.

KEYWORDS: Cryocooler, Joule-Thomson, microcryocooler, micro-JT, mixed refrigerants.

Advances in Cryogenic Engineering
AIP Conf. Proc. 1434, 690-697 (2012); doi: 10.1063/1.4706980
2012 American Institute of Physics 978-0-7354-1020-6/$0.00

INTRODUCTION

Micro-scale cryogenic coolers are fast becoming of great interest, as they are intended for low power-consumption sensors such as new-generation mid-wavelength infrared (IR) sensors and enabling superconductivity in some sensors. Compact size and stable temperature reduces thermal noise and input power for both sensors and MCCs. In the 1990s, Little [1- 2] discussed potential for MCCs to cool electronic chips and other devices. He developed J-T MCC's based upon etched glass plate heat exchangers that were about the size of a matchbox using high-pressure nitrogen as the refrigerant. He also discussed use of mixed refrigerants to have potential for MCC's.

While Marquardt *et al.* [3] successfully employed a mixed-gas J-T MCC for medical applications in the late 1990s, Burger *et al.* [4] in the 2000s presented a micro-machined cryocooler employing a counter flow heat exchanger made by inserting a single glass capillary (0.25 mm ID, 0.36 mm OD) into a larger one (0.53 mm ID, 0.67 mm OD), forming a coaxial heat exchanger that resulted in a 77 mm x 9 mm MCC employing ethylene at up to 20:1 bar pressure ratio. Later in the 2000s, Lerou *et al.* [5-6] demonstrated microfabricated cryocoolers with 30 mm x 2.2 mm glass-plate-based J-T MCCs employing nitrogen at 80:6 bar pressure ratio. More recently, Bradley *et al.* [7] presented a microfabricated cooler with 25 mm long multichannel glass fiber heat exchanger of 6 high-pressure channels ID/OD=75 μm/125 μm inserted in a single low-pressure capillary ID/OD=536 μm/617 μm with a microelectromechanically fabricated expansion valve (0.760 μm tall x 500 μm long radial restriction) employing a 5-component mixed refrigerant at 16:1 bar low pressure ratio.

Stable and distributed temperature operation is a hallmark for J-T coolers. However, temperature fluctuations for J-T cryocoolers are not unheard of. The advent of micro-scale J-T cryocoolers brings the potential for undesirable temperature instability due to fixed micro-scale expansion valves and heat exchangers in collaboration with the employment of mixed-refrigerants. As the scale of the cryocooler decreases, so does the stabilizing mass at the cold tip. With low masses, on the order of low tens or even single grams down to milligrams, stable temperature operation with very low excursion of ≤ 1 K without active means of stabilization presents some challenge.

Longsworth [8], Luo [9], and Maytal [10] all make reference to fluctuations in temperature whether from flow fluctuations or intermittent clogging. Solutions have come by way of varying the J-T valve restriction during operation in some manner to alleviate the restriction to reestablish proper flow and/or remove clogs, which take place primarily at or near the J-T expansion valve. While this has proven to be effective for moderate scale J-T coolers (and larger), it is not a readily employable technique for micro-scale J-T coolers, as the inherent size limits J-T expansion valves to a fixed restriction in addition to flow passage area within the heat exchanger.

Recent test results for a 5-component hydrocarbon mixture operated at 14:0.7 bar pressure ratio in an MCC of the type referred to by Bradley *et al.* [7] demonstrated pseudo-stable temperatures (140 K, 150 K, and 168 K) and rapid temperature decreases (drops to 76 K and 85 K) with periods of widely fluctuating temperatures. Such behavior is indicative of flow fluctuations with intermittent clogging and/or phase changes from liquid to vapor. Considering the scale and design, fluctuations of this nature are not entirely unexpected as intermittent liquid to vapor flow likely exists in the cold head during operation (FIGURE 1 shows details and configuration for the MCC: counterflow heat exchanger with exterior segmented gold plating that is 25 mm tall with a 2 x 2 x 1 mm silicon and glass cold tip encapsulating the 760 μm tall microvalve.) FIGURES 2 and 3 illustrate these temperature behaviors quite well for a micro-scale J-T cryocooler.

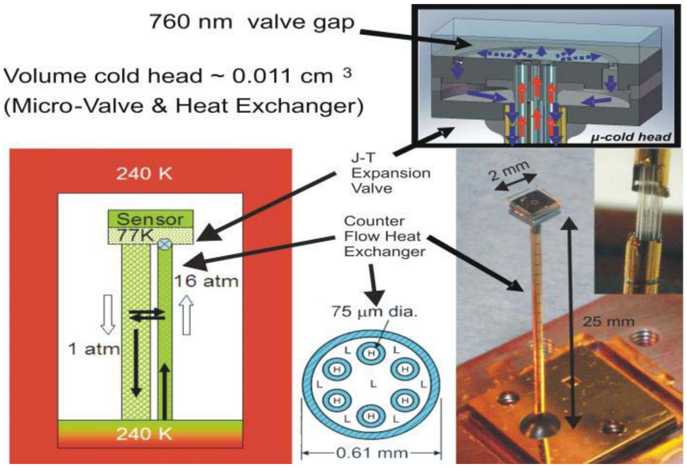


FIGURE 1. MCC Clockwise from lower left; MCC schematic, micro-valve cold tip, close up of high- and low-pressure fibers in heat exchanger, complete cold head assembly, and heat exchanger cross-section.

MICRO AND MESO-SCALE J-T

For J-T systems, gross refrigeration is the product of the molar flow rate $\dot{n}$ and the minimum enthalpy difference, $(\Delta h)_{\min}$ of the high and low-pressure enthalpies for the temperature range of interest, given by

$$\dot{Q} = \dot{n} \bullet (\Delta h)_{\min} . \tag{1}$$

Employing a gas mixture refrigerant serves to improve the J-T refrigeration by improving the minimum enthalpy difference between the high- and low-pressure enthalpies that occur during expansion, thereby improving the gross refrigeration for a given flow rate and ratio of inlet to outlet pressures. FIGURE 4 shows the minimum enthalpy for the 5-component mixture (14 % propane, 16 % ethane, 22 % methane, 42 % nitrogen, and 6 % neon by mole fraction) optimized over the range of 76 K to 240 K [11-13] employed in the 16:1 bar aforementioned test. Precooling to temperatures lower than ambient serves to establish the greatest value for minimum enthalpy difference, $(\Delta h)_{\min}$ over the temperature range of operation for a desired low temperature. It should be noted that while the pressure ratio of 16:1 bar is easily achieved for other J-T cryocoolers, it is too high for

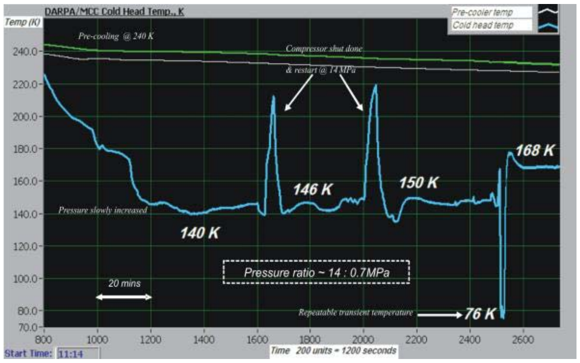


FIGURE 2. Stable temperature with 16:1 ratio mixed refrigerant.

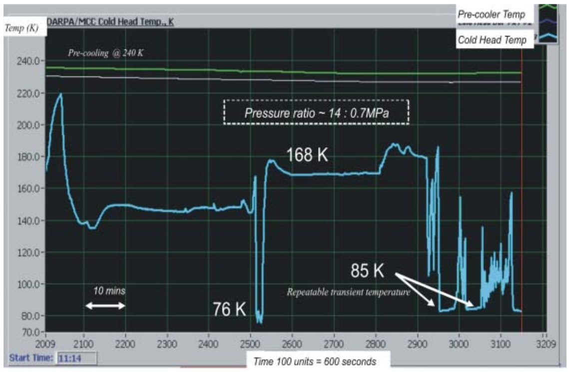


FIGURE 3. Rapid temperature changes for 16:1 mixed refrigerant.

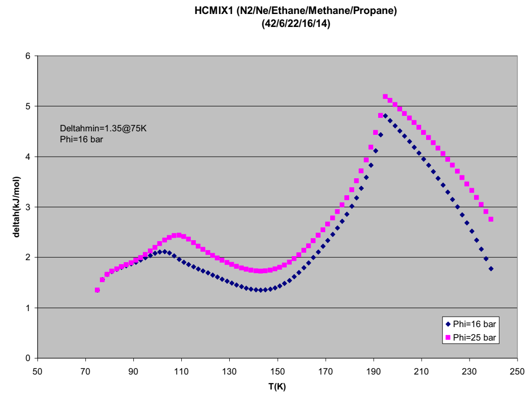


FIGURE 4. Enthalpy difference for 16:1 ratio mixed refrigerant (dark markers).

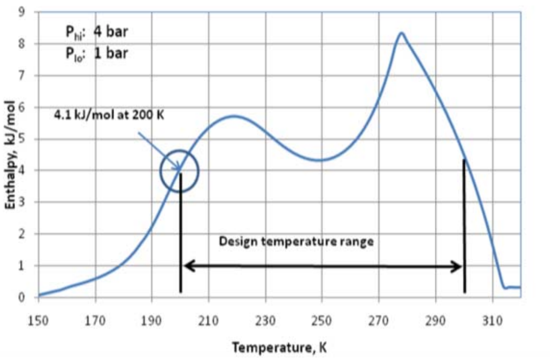
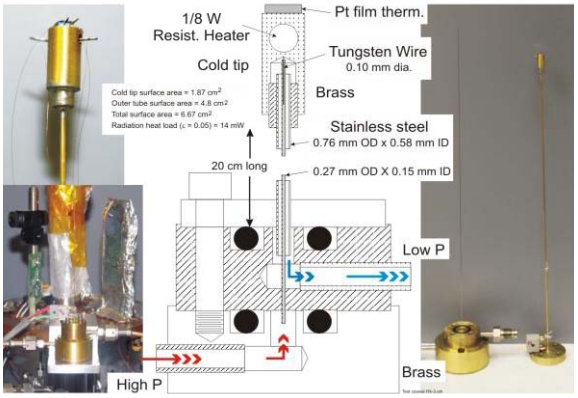


FIGURE 5. Enthalpy difference for 4:1 ratio mixed refrigerant.

miniature, let alone micro-scale, compressors at this time. Thus, by readjusting to a more moderate 4:1 bar pressure ratio, a given refrigeration may be achieved for a flow rate that compensates for $(\Delta h)_{\min}$ values, thereby allowing operating pressures more suitable for micro-scale compressors. A 5-component mixture refrigerant optimized over the range of 200 K to 300 K [11-13] composed of 8% methane, 46% ethane, 14% propane, 4% butane, and 26% pentane was arrived at that provides $(\Delta h)_{\min} = \sim 4.0$ kJ/mol, at 200 K shown in FIGURE 5. The MCC cold head is the same as mentioned previously but with a taller valve gap of 1.8 μm to accommodate a lower pressure of 4 bar for an expected flow of ~ 10 cm^3/min.

Observing temperature fluctuations during tests with a 16:1 mixture refrigerant, we concluded that a meso-scale version of the cooler should be explored to evaluate gas mixtures and temperature instabilities that may arise, particularly for low ratio 4:1 bar mixtures. Thus, an easily modified meso-scale cold head (as shown in FIGURE 6) was constructed for a higher flow of ~ 30 cm^3/min. compared with the design flow of ~ 10 cm^3/min. for the MCC. The meso-scale J-T here is a simple design which employs a single high-pressure tube (ID/OD=100μm/266μm) inserted within a single outer low pressure tube (ID/OD=58 μm/760 μm) to form the counter flow heat exchanger that is 20 cm long. The J-T expansion valve is formed by use of a tungsten wire (OD=100 μm) inserted 11 mm into the high pressure line at the cold tip which encapsulates the valve and expansion volume in a brass cap. The exterior of the heat exchanger and the cold tip are gold plated to minimize radiation loss. The J-T valve impedance (insertion depth for the tungsten



Cold tip surface area = 1.87 cm^2
Outer tube surface area = 4.8 cm^2
Total surface area = 6.67 cm^2
Radiation heat load (ε = 0.05) = 14 mW

FIGURE 6. Meso-scale cooler clockwise from upper left; Cold tip, cooler schematic, cold head assembly, and cold head in test apparatus.

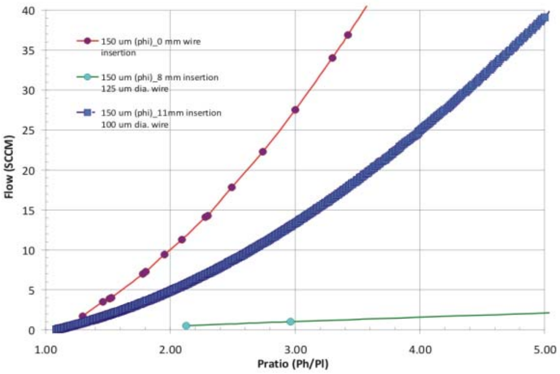


FIGURE 7. Flow curves to determine impedance for the meso-scale cooler with nitrogen.

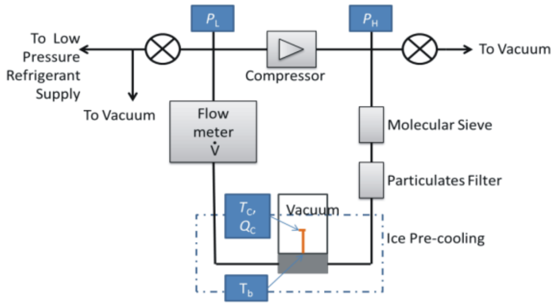


FIGURE 8. MCC test setup with ice precooling mixture to ~ 275 K.

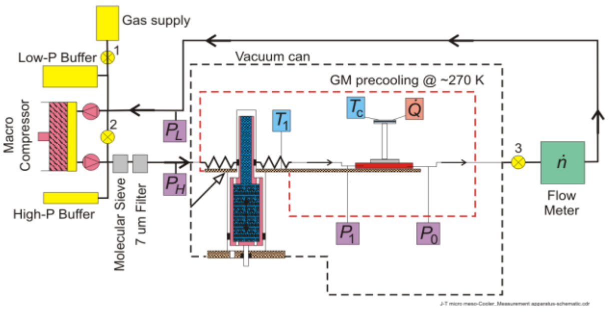


FIGURE 9. Meso-scale test setup with GM cryocooler providing precooling of mixture to ~ 270 K.

wire) is determined directly from measurements of flow versus pressure across the heat-exchanger and expansion valve assembly using nitrogen gas, as indicated in FIGURE 7.

MCC AND MESO-SCALE TESTS WITH 4:1 MIXTURE

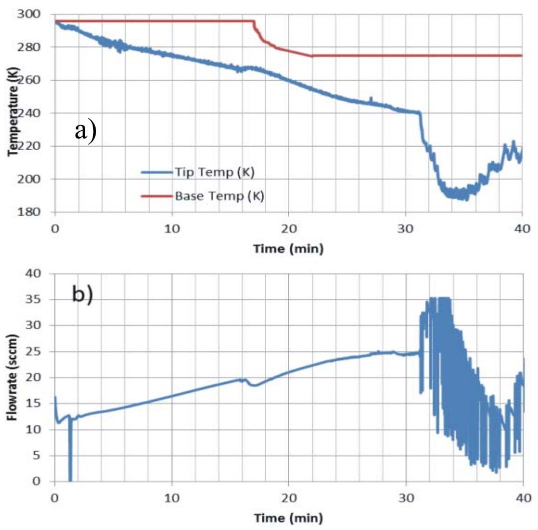
MCC and Meso-scale Test Setup

The test setup for the MCC and the meso-scale coolers are quite similar, but with two important differences. The MCC setup utilizes a miniature non-lubricated compressor with MEMS fabricated check valves and employs ice to precool to ~ 275 K (see FIGURE 8). Further discussion of the system performance is presented by Lewis *et al*. [14]. Whereas the meso-scale setup utilizes a macro-scale linear drive non-lubricated compressor and utilizes a Gifford-McMahon (GM) cryocooler to precool to the desired temperature, ~ 270 K in this instance (see FIGURE 9). Trace levels of water leading to clogging issues in microcryocoolers have been documented by Lerou *et al*. [15] with obvious understanding that any/all trace contaminants migrate to the lowest temperature region of all cryocoolers. Thus both setups employed 3 Å (1 g for MCC, 4 g for meso-scale) molecular sieve to dry the mixture with particulate filters (15 µm for MCC, 7 µm for meso-scale) just upstream of entry to the cold head. The 4:1 bar mixture was custom made by a local gas supplier to be dry (free from water) and free of particulate contaminants for an optimized temperature range of 200 K to 300 K with precooling to about 270 K (see FIGURE 5).

MCC and Meso-scale Temperature Instabilities – Measurements and Discussion

Evaluations were made for both the MCC and the meso-scale coolers employing the 5-component mixture consisting of 8 % methane, 46 % ethane, 14 % propane, 4 % butane, and 26 % pentane with precooling to about 270 K in each case with dryer and filters in place during testing. Temperature instabilities characterized by periodic fluctuations in temperature and flow were observed for both coolers. Sizeable drifts in temperature and flow were observed for the MCC alone.

Employing the MCC test setup cooling of the MCC is shown in FIGURE 10. There are some fluctuations of a few kelvins or more that coincide with sizeable fluctuations in flow similar to that exhibited with the 16:1 bar low-pressure mixture (review FIGURES 1 and 2). For the MCC, rapid cooling adjoins rapid increase in flow while warming adjoins decrease in flow accordingly as viewed during the 30 to 40 minute timeframe. We also observe that minor decrease in flow of about 3 SCCM corresponds to an increase in temperature of about 4 K during the 16 to 18 minute time-frame. Rapid swings of 10 to 20



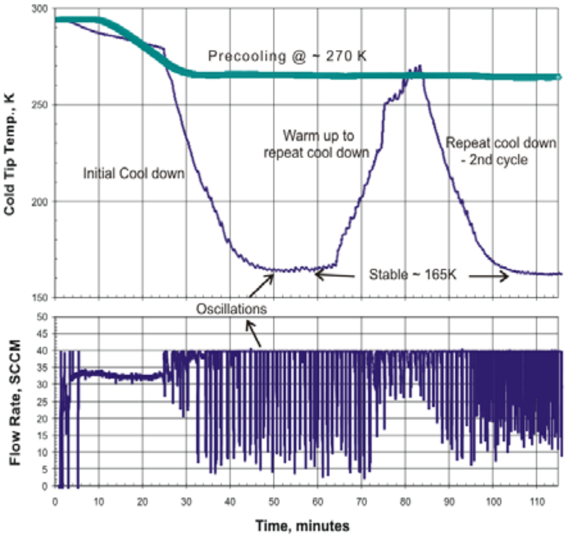
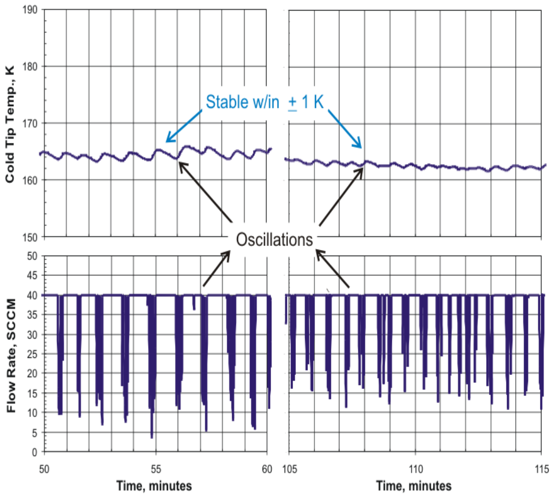


FIGURE 10. MCC cool down with ice precooling. Flows give rise to temp. fluctuations & instability.

FIGURE 11. Meso-scale cool down w/GM cryo-cooler. Flow gives rise to temp. fluctuations, but stable temp. achieved

SCCM, particularly decreases, lead to rapid temperature fluctuations. The temperature fluctuations are symptoms of the irregular/intermittent flow within the cold head. This is easily observed and should be expected for the MCC, as the cold tip mass is quite low, ~ 8.5 mg, and has a very low radiation load of ~ 1.4 mW. Thus, there is very little mass to dampen the magnitude of these, or the larger, fluctuations exhibited during the 30-to-40 minute timeframe of the test. It becomes apparent that the general trend in flow, whether it be increasing or decreasing, leads to a similar trend in temperature. It is interesting to note the apparent-stair stepping in the temperature as the MCC cools or as it warms after bottoming out.

The meso-scale test setup was employed to test the meso-scale cooler for similar operating conditions of the MCC test. With precooling just below 270 K, the meso-scale cooler also exhibits temperature fluctuations. In FIGURE 11 the fluctuations become more obvious during cool down as the temperature approaches 200 K. Upon closer inspection (view FIGURE 12), the temperature fluctuations correspond with the fluctuations in flow, which are repeatable and periodic, with reoccurrences about every 1.5 minutes (longer period than that of the MCC). This is consistent, as the meso-scale cold tip mass is ~ 1750



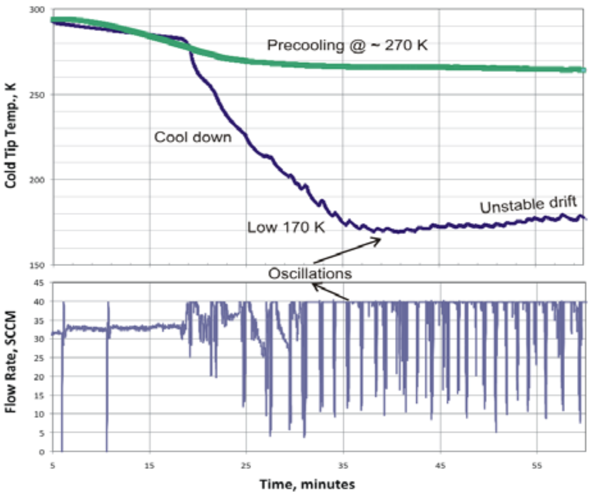


FIGURE 12. Closer inspection of flow and temp. fluctuations of meso-scale cooler test (w/dryer).

FIGURE 13. Meso-scale cool down w/o molecular sieve dryer. Unstable low temp. that drifts.

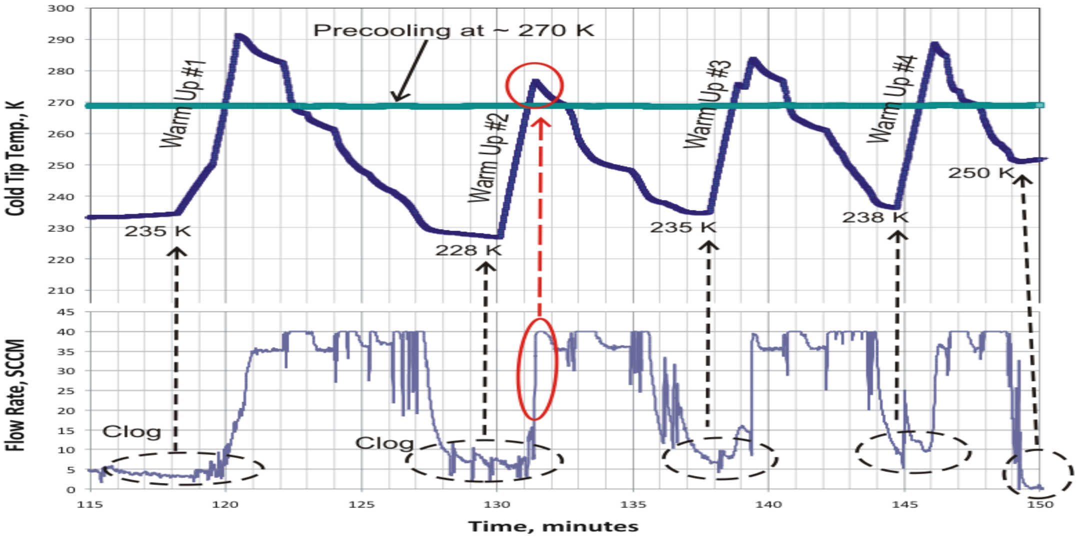


FIGURE 14. Without molecular sieve, meso-scale cooler exhibits numerous repeated clogs.

mg and has a radiation loss of ~ 14 mW. The general trends for both flow and temperature are evident much the same as for the MCC, although with good long-term general temperature stability (little drift).

Contaminants are important considerations for MCCs, and particularly for water that may be entrained in mixed refrigerants. Thus, another test of the meso-scale cooler was conducted under the same operating conditions. However, the molecular sieve dryer was omitted, eliminating any drying of the mixture, with interesting results.

The initial cool down was similar to that with molecular sieve drying (view FIGURE 13), exhibiting similar minor flow and temperature fluctuations during cool down. However, after a lengthy period of operation the cold tip began to experience clogging, exhibited by a decrease in flow with a resulting increase in temperature as shown in FIGURE 14. Upon warm up to above 275 K, the flow returns to $\geq$ 30 cm^3/min, and after removing heat input the cold tip returns to a low temperature of only ~ 230 K, whereupon the clogging reestablishes. Subsequently, during repeated cycles of cool down and heat up, clogging repeated at increasingly higher temperatures of up to ~ 250 K, suggesting a build-up of contamination that could not be completely removed at the cold tip.

The temperature at which flow returns to normal is indicative of water (freezing at the cold tip of course), albeit in extremely low amounts within the mixture (the mixture was procured commercially to be dry). Thus, previous use of molecular sieve to trap residual amounts present in the mixture proved successful. Maytal [10] and Lerou [15] discussed clogging resulting from trace amounts of water in the refrigerant whether a mixture or pure fluid. These results emphasize need for drying and/or preconditioning of mixed refrigerants for micro-scale J-T coolers.

CONCLUSION

Flow instabilities leading to temperature fluctuations and drift have been shown to exist for both micro and meso-scale J-T cryocoolers employing mixed refrigerants operating at low pressure ratios of 4:1 bar. Contaminants in the refrigerant, such as water, represent significant issues for stable temperature and long term operation of any MCC. The MCC, having been designed for mixed refrigerants to operate with flows of about 6 - 10 cm^3/min has capably demonstrated cooling to below 200 K at close to 30 cm^3/min but with fluctuations in flow that give rise to temperature fluctuations that lead to upward drift in temperature thereby, losing stability. The drift and instability in temperature arise from the unstable flow exacerbated by low mass at the cold tip. In comparison, the meso-scale cooler designed for similar refrigerants with flows of about 3 to 5 times those of the MCC design has demonstrated fluctuations in flow that give rise to temperature fluctuations of a few kelvin with a periodicity of ~ 1.5 minutes but remains very stable at a low temperature of ~ 170 K. While increased mass at the cold tip aids to dampen the resultant fluctuations, they are not eliminated, suggesting that the meso-scale cooler does present opportunities for characterizing and validating mixed refrigerants for micro-scale coolers. However, as the scaling of flow passages and flow rates within the J-T valve and heat exchanger decrease there is optimism that we have yet to reach the threshold level that inhibits the potential of mixed refrigerant use in MCCs. We simply have more work to do to overcome these obstacles.

ACKNOWLEDGMENTS

We gratefully acknowledge support of this work by the DARPA Micro Cryogenic Cooler Program.

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