Joule-Thomson Cryocoolers Operating with Binary Mixtures

N. Tzabar

Rafael
Haifa, Israel 3102102

ABSTRACT

Joule-Thomson (JT) cryocoolers operating with mixtures containing 4 to 7 components are widely investigated for reducing the operating pressure and increasing the COP. In addition to exploring multi-component mixtures, we also examine mixtures with 2 to 3 components as refrigerants for JT cryocoolers. Though the performance of these mixtures is expected to be inferior relative to multi-component mixtures, their performance can be analytically calculated in a more convenient manner. Furthermore, in some scenarios, binary and ternary mixtures can be advantageous over multi-component mixtures.

In previous research we have investigated phase diagrams and isothermal JT effects (Δh_T) of binary mixtures that contain nitrogen, argon, methane, ethane, ethylene, and propane. The present paper presents our research progress with nitrogen-ethane and nitrogen-propane mixtures for the extended cooling temperature range of 80 – 150 K. Compression power is calculated and the COP of the cryocooler is obtained. Experimental results are presented to verify some of the analytical results.

INTRODUCTION

JT cryocoolers operating with pure gases provide steady cooling temperatures without any active control and benefit from performance that is independent of ambient temperature. Still, mixed refrigerants (MR) for Joule-Thomson (JT) cryocoolers have been widely investigated for various cooling temperatures due to two main advantages: lower pressure ratios and higher coefficient of performance (COP). Multi-component mixtures are usually in use in order to obtain enhanced performance [1-4]. Since binary mixtures are not the optimum refrigerants, they are rarely considered as refrigerants; however, they are analytically investigated in a more convenient manner. Xu et al. [5,6] examined flammable and non-flammable binary mixtures for Joule-Thomson cryocooling applications. Tzabar and Grossman [7] investigated several types of binary mixtures as refrigerants for cooling to 80 – 100 K. Although data of the mixtures is available in the literature [8,9] the ability of calculating the phase diagram [10] is required in order to investigate MRs.

The current paper presents research on JT cryocoolers operating with nitrogen-ethane and nitrogen-propane MRs. The operating pressures for cooling temperatures between 80 K and 150 K are determined. The COPs of the different refrigerants are compared and experimental results are presented to verify the calculated results.

Cryocoolers 18, edited by S.D. Miller and R.G. Ross, Jr.

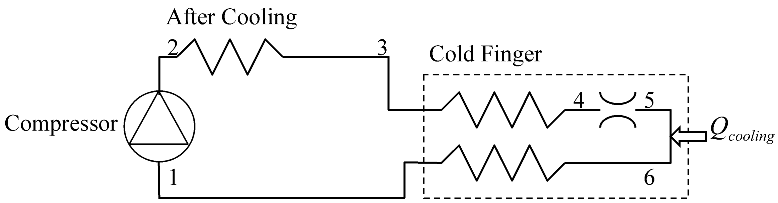

Figure 1. A schematic of the Joule-Thomson cryocooler.

METHOD OF STUDY

Cooling Temperature

A schematic of the cryocooler is presented in Figure 1. This is a basic configuration of a JT cryocooler with a single compressor, a passive aftercooler that reduces the compressed fluid temperature to the ambient temperature, and a Linde-Hampson type cold-finger. The required cooling temperature of a given mixture is calculated according to the liquid line of the phase diagram. In a previous work [10] several methods for calculating the liquid line were examined and the γ-ϕ method has been found preferable. This method was further used in an earlier work on binary mixed-refrigerants for JT cryocoolers [7].

Additional demand for obtaining cooling temperatures that equal the liquefaction temperatures is to have a sufficient specific cooling power (isothermal JT effect, Δh_T) for the entire temperature range of interest, in other words, to avoid pinch points along the enthalpy-temperature plots. The enthalpies of the mixtures are calculated with REFPROP™ software. The mixture compositions are listed in Table 1.

Coefficient of Performance (COP)

The COP is calculated by:

$$COP = \frac{Q_{cooling}}{W_{compression}} \tag{1}$$

$$Q_{cooling} = \dot{m}(h_1 - h_3) \tag{2}$$

$$W_{compression} = \frac{\gamma}{1-\gamma}\dot{m}\frac{R}{M}T\left[\left(\frac{p_2}{p_1}\right)^{\frac{\gamma-1}{\gamma}} - 1\right] \tag{3}$$

where $\dot{m}$ is the flow rate, γ is the heat capacities ratio, R is the universal gas constant, and M is the molecular weight. T is temperature, h is the enthalpy, and p is the pressure. All the numbered subscripts refer to the states in Figure 1. One should notice that the COP does not depend on the flow rate; thus, the specific cooling power is discussed.

Table 1. Mixture Compositions in Mole Fractions.

	nitrogen	ethane	propane
MIX-1	0.4	0.6	-
MIX-2	0.55	0.45	-
MIX-3	0.7	0.3	-
MIX-4	0.6	-	0.4

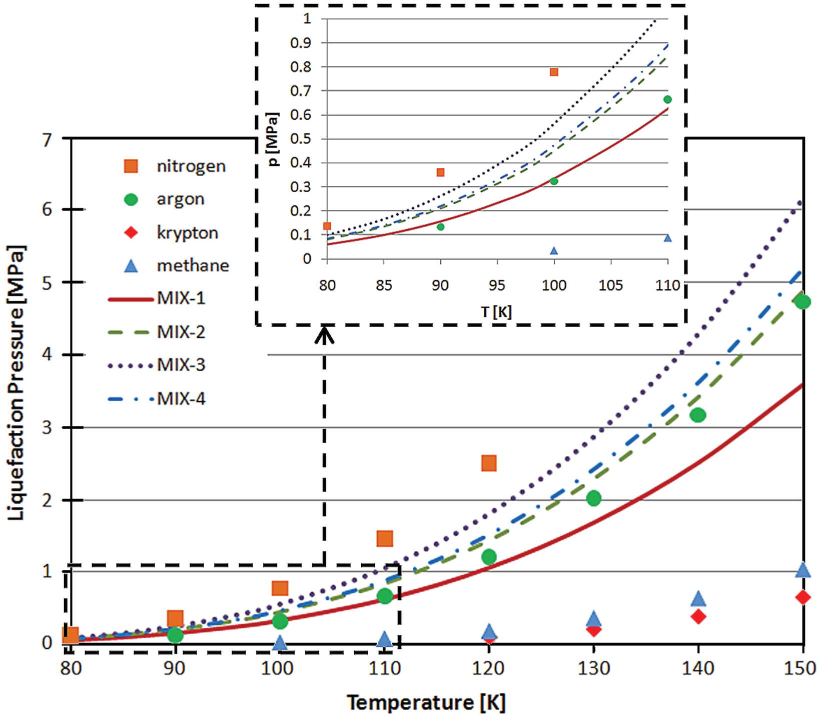

Figure 2. The required pressure, according to phase equilibrium considerations, of several pure and mixed refrigerants for liquefaction at temperatures between 80 K and 150 K.

Experimental Apparatus

Experiments with the four mentioned binary MRs were conducted. The experimental apparatus includes an oil-free laboratory compressor, a JT cold-finger, a temperature diode for measuring the cooling temperature, and two pressure transducers at point 1 and point 2 (see Figure 1). The cold-finger is located in a temperature chamber that controls its ambient temperature and the temperature of the incoming fluid, T_3. Two types of binary mixtures are examined; nitrogen-propane and nitrogen-ethane.

RESULTS AND DISCUSSION

Cooling Temperature

Figure 2 shows the required pressure of several pure and mixed fluids to obtain liquefaction temperatures between 80 K and 150 K. Cooling to 80 K requires about 0.1 MPa for all fluids that are suitable for this liquefaction temperature. Increased liquefaction temperatures expose the differences among the fluids; a pure fluid with higher normal boiling temperature requires lower pressure for a given liquefaction temperature, nitrogen-ethane and nitrogen-propane mixtures require similar pressures, and an increase of the hydrocarbon component in the mixtures reduces the required pressure for a given liquefaction temperature.

In order to obtain the liquefaction temperature as the cooling temperature, pinch point at the isothermal JT effect shall be avoided. Figure 3 shows the isothermal JT effect of three nitrogen-ethane mixtures; MIX-1, MIX-2, and MIX-3, and a single nitrogen-propane mixture; MIX-4. The results in Figure 3 are for a cooling temperature of 100 K and a high pressure of 2 MPa. The low pressure for each mixture is detailed in the figure. Pure refrigerants have their lowest isothermal JT effect at the highest temperature of interest; therefore, cooling down to the liquefaction temperature

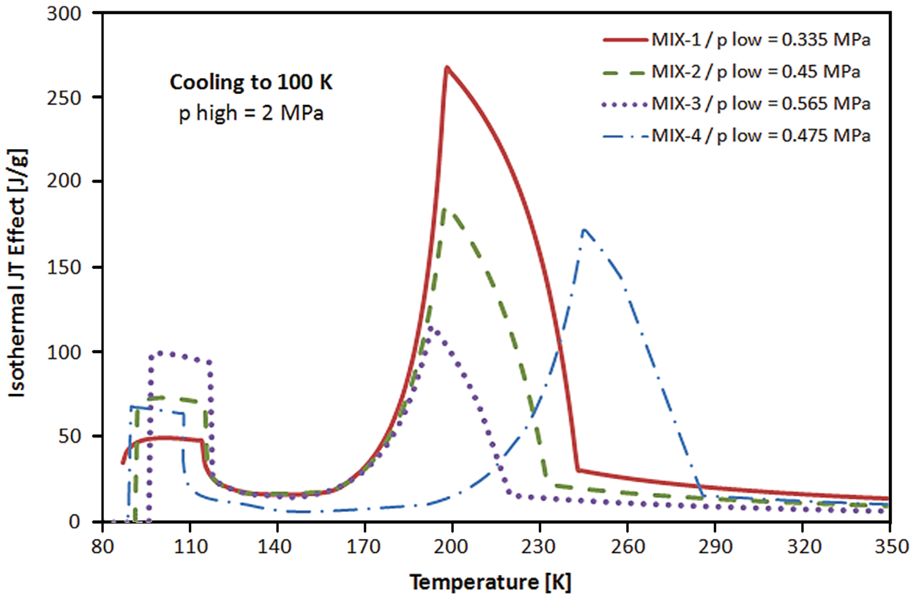

Figure 3. Isothermal JT effect of nitrogen-ethane mixtures with three compositions. The indicated low pressure of each mixture suits a cooling temperature of 100 K and the high pressure is constant and equals 2 MPa.

is feasible. MRs have a more complicated isothermal JT effect as demonstrated in Figure 3. All nitrogen-ethane mixtures have a decrease in their isothermal JT effect around 150 K to the same value; however, for MIX-1 it is a pinch point that interrupts the cooling process, while for MIX-3 it isn't. MIX-2 has approximately a balanced composition where the pinch points appear. The results of MIX-4 demonstrate the pinch point that was obtained in all the nitrogen-propane binary mixtures that were examined.

Coefficient of Performance (COP)

The COPs of the mixtures were examined against several operating parameters. Figure 4 shows the COPs versus the required pressure ratio for cooling temperatures from 80 K to 150 K where liquefaction is attained. At every cooling temperature the relevant coolants are shown. The results are presented as a function of the pressure ratio, since it defines the required compressor, number of compression stages, input power, and compressor availability. Figure 4 proves that MRs have improved COP compared to pure refrigerants. Increase in the pressure ratio increases the COP; however, the rate is larger for MRs than for pure refrigerants. At a cooling temperature of 80 K, MIX-1 has a performance similar to MIX-2. MIX-2 has better COP compared to MIX-3, but is only suitable for cooling temperatures between 80 K and 100 K, while MIX-3 is suitable for cooling temperatures between 80 K and 140 K. MIX-4, which contains nitrogen and propane, has the highest COP and is feasible for cooling temperatures between 80 K and 90 K.

Experimental Results

Many experiments with the four mixtures have been conducted. Figure 5 demonstrates an experiment with MIX-1 where the cooling temperature is maintained steady and equal to 92 K. The low pressure is 0.1 MPa and the high pressure is 2.7 MPa. Figure 6 shows another experiment with MIX-1 and shows the relation between the cooling temperature and the operating pressures, espe-

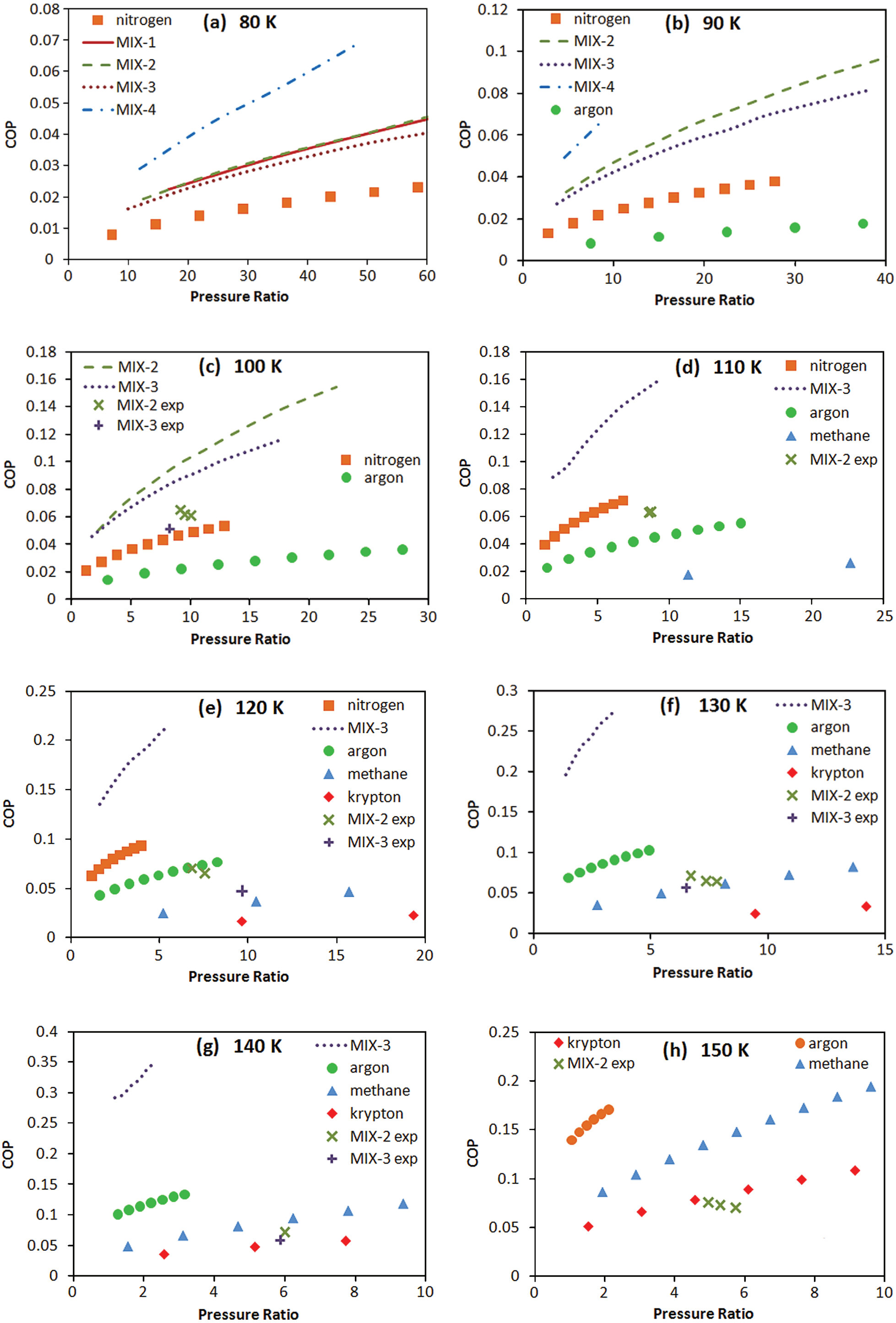

Figure 4. Calculated COP as a function of the pressure ratio for different fluids at various cooling temperatures. Several experimental results of the mixtures are also presented.

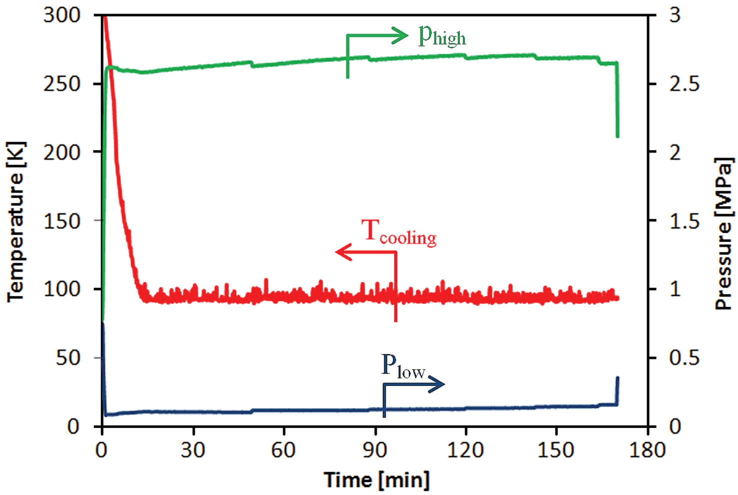

Figure 5. Steady cooling temperature operation with MIX-1.

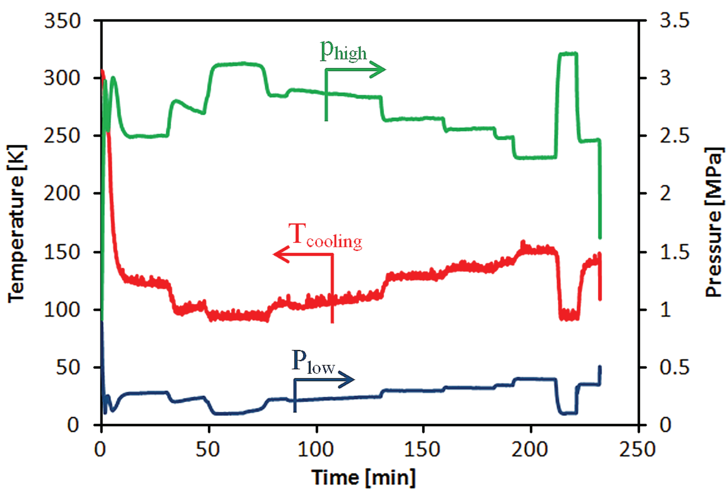

Figure 6. Experimental results with MIX-1 demonstrating the control of the cooling temperature by changing the operating pressures.

cially the low pressure. The results demonstrate controlling the cooling temperature by changing the operating pressure of the compressor. The cooling temperature is varied between 90 K and 150K by varying the low pressure between 0.1 MPa and 0.4 MPa, respectively. The high pressure is varied between 2.4 MPa and 3.3 MPa, which provides the required cooling power. The results in Figure 6 prove the repeatability of the cooling system, meaning that the operating conditions are stable. Figure 7 shows experimental results with MIX-2 at ambient temperatures between 290 K and 340 K. The cooling temperature remains steady at 105 K while the low pressure is changed between 0.2 MPa and 0.4 MPa and the high pressure is varied between 2.85 MPa and 3.6 MPa.

Figure 4 also presented some experimental results against the calculated results. As expected, the practical COP is lower than the calculated COP; this is best shown in Figure 4 (c) for MIX-2 and MIX-3 at a cooling temperature of 100 K. MIX-3 at a pressure ratio of 8.3 has experimental and calculated COPs of 0.051 and 0.082, respectively. And MIX-2, at a pressure ratio of 9.6, has

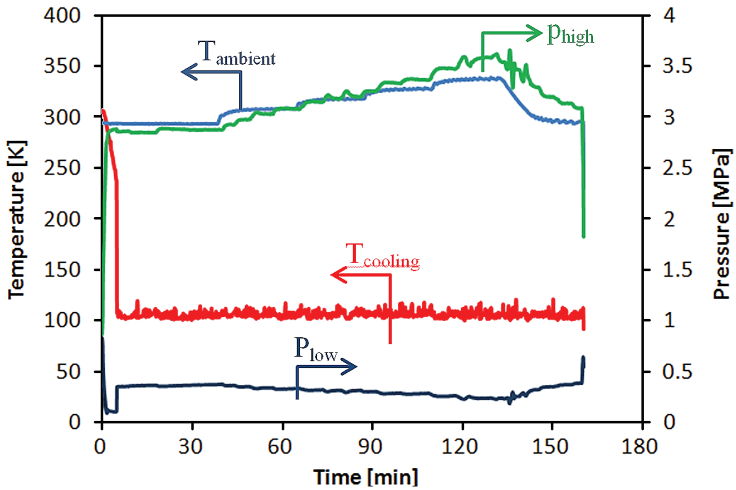

Figure 7. Experimental results with MIX-2 at ambient temperature between 290 K and 340 K and steady cooling temperature that equals 105 K.

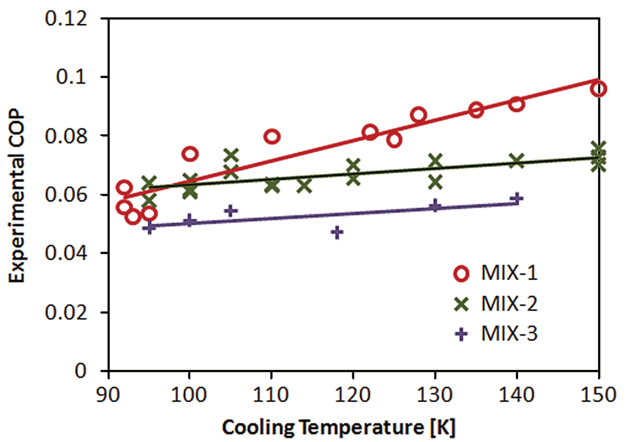

Figure 8. Experimental COP of MIX-1, MIX-2, and MIX-3 versus the cooling temperature.

experimental and calculated COPs of 0.062 and 0.1, respectively. Both experimental results are 62% of their calculated estimations. The deviations between experimental and calculated results are more severe for higher cooling temperatures, since the current experimental apparatus can't provide the required operating pressures for cooling temperature above 100 K; therefore, these experiments are at non-optimal operating conditions. In addition, this is the reason for presenting experimental results in figures that don't show calculated results.

The experimental results of MIX-1, MIX-2, and MIX-3 at normal ambient temperature (about 295 K) are summarized in Figure 8 where the COP is shown versus the cooling temperature. Linear trend lines are added to assist interpreting the results. These results show that MIX-1 has the highest COP and MIX-3 has the lowest COP due to the concentration of ethane in the mixture. The COP of all mixtures increases when the cooling temperature increases, as expected. As mentioned above, in the current experiments, cooling to temperatures above about 100 K is at non-optimal conditions; therefore, if a more appropriate apparatus was used, better COPs could be expected for these cooling temperatures.

CONCLUSIONS

The present paper describes calculated and experimental results for JT cryocoolers operating with nitrogen-ethane and nitrogen-propane mixtures. The results show the ability of estimating the cooling temperatures for binary mixtures and their COPs which are advantageous over pure refrigerants. It is desired to obtain liquefaction of the mixture to achieve the benefits of a steady cooling temperature; however, the experimental results show that it is possible to attain steady cooling temperatures without obtaining complete liquefaction of the working fluid at the expense of lower cooling power and COP.

Nitrogen-ethane mixtures show more stable performance relative to nitrogen-propane mixtures; therefore, they are recommended for use. A higher concentration of ethane in the mixture provides better COP; however, it also limits the range of feasible cooling temperature.

Steady cooling to temperatures between 90 K and 150 K with a single cryocooler and nitrogen-ethane mixture is demonstrated. Also demonstrated is steady temperature operation at ambient temperatures between 290 K and 340 K. Cooling to temperatures above 100 K while attaining liquefaction of the working fluid isn't obtained, since the existing experimental apparatus was not capable of providing the required operating pressures. These results show the advantage of binary mixed refrigerants that, at the appropriate conditions, function in a similar manner to pure refrigerants with enhanced specific cooling power.

REFERENCES

1. Nayak H.G. and Venkatarathnam G., "Performance of an auto refrigerant cascade refrigerator operating on gas refrigerant supply (GRS) mode with nitrogen-hydrocarbon and argon-hydrocarbon refrigerants," *Cryogenics*, vol.49 (2009), pp. 350-359.

2. Walimbe N.S., Narayankhedkar K.G., and Atrey M.D., "Experimental investigation on mixed refrigerant Joule-Thomson cryocooler with flammable and non-flammable refrigerant mixtures," *Cryogenics*, vol.50 (2010), pp. 653-659.

3. Khatri A. and Boiarski M., "Development of JT coolers operating at cryogenic temperatures with nonflammable mixed refrigerants," *Advances in Cryogenic Engineering - CEC*, vol. 53(2008), pp. 3-10.

4. Tzabar N. and Lapp Z., "Experimental investigation on mixed refrigerant for closed-cycle Joule-Thomson cryocoolers," *Advances in Cryogenic Engineering - CEC*, vol.55 (2010), pp. 1121-1128.

5. Xu M., He Y., and Chen Z., "Analysis of using binary cryogenic mixtures containing nitrogen and alkanes or alkenes in cryocoolers," *Cryogenics*, vol.36 (1996), pp. 69-73.

6. Xu M., He Y., and Chen Z., "Analysis of using binary cryogenic mixtures containing nitrogen and freon in cryocoolers," *Cryogenics*, vol.36 (1996), pp. 243-247.

7. Tzabar N. and Grossman G, "Binary mixed refrigerants for Joule-Thomson cryocooling to 80-100 K," *Cryocoolers 17*, ICC Press, Boulder, CO (2012), pp. 387-296.

8. Grauso L., Fredenslund A., and Mollerup J., "Vapour-liquid equilibrium data for the system C_2H_6+N_2, C_2H_4+N_2, C_3H_8+N_2, and C_3H_6+N_2," *Fluid phase equilibria*, vol.1 (1977), pp. 13-26.

9. Carrero-Mantilla J. and Llano-Restrepo M., "Vapor-liquid equilibria of the binary mixtures nitrogen + methane, nitrogen + ethane and nitrogen + carbon dioxide, and the ternary mixture nitrogen + methane + ethane from Gibbs-ensemble molecular simulation," *Fluid phase equilibria*, vol.208 (2003), pp. 155-169.

10. Tzabar N., "Phase equilibria in binary mixtures at cryogenic temperatures," *Proceedings of the ICEC24-ICMC 2012*, Fukuoka, Japan, May 14-18, 2012, pp. 177-180.

11. Stanley IS., Chemical, biochemical, and engineering thermodynamics 4th ed, John Wiley & Sons Inc., USA (2006).