Experimental investigation on mixed refrigerant Joule–Thomson cryocooler with flammable and non-flammable refrigerant mixtures

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ABSTRACT

The mixed refrigerant Joule–Thomson (MR J–T) cryocoolers have a wide application area covering the temperature range from 80 K to 200 K. The significant advantages of the system are simplicity of its design and working reliability with high level performance. The present paper discusses the experimental results of MR J–T cooler with different flammable and non-flammable mixture compositions. The work highlights the use of pressure–enthalpy and temperature–enthalpy diagrams for these mixtures to support the experimental results. A record lowest temperature of 65 K and a cooling capacity of 6 W at 80 K are obtained for a single stage MR J–T system starting at 300 K. Further, using a mixture of minimum flammable refrigerants, temperatures below 100 K is achieved.

1. Introduction

In recent past, there has been significant development in closed cycle mixed refrigerant Joule–Thomson (MR J–T) cryocoolers. The use of mixtures in Joule–Thomson (J–T) coolers is justified by Brodyanskii et al. [1]. The gas mixtures used in MR J–T coolers have mainly very low boiling components such as helium, neon, nitrogen, argon and light hydrocarbons ranging from methane to pentane. MR J–T systems, working with typical high and low pressure limits of 2.0–3.0 MPa and 0.1–0.4 MPa respectively, have been discussed in the literature [2–7]. The system has many advantages such as easy fabrication, no critical machining of any parts, comparable cooling effect with other cryocoolers, moderate pressure ratio, minimum vibrations, high reliability and less cost. Hence the closed cycle MR J–T system has many ground based applications covering wide temperature range of 80–200 K. One of the relatively high temperature applications (120–200 K) is in the medical field where the mixture should preferably consist of non-flammable components due to safety reasons. Podtcherniaev et al. [8] have reported the performance of single and two stage MR J–T coolers using hydrofluorocarbon components for high temperature applications. Boiarski et al. [9] have presented a performance comparison between non-flammable refrigerant mixtures and flammable refrigerant mixtures. Khatri and Boiarski [10] reported experimental and modeling work related to single stage MR J–T cooler for the temperature range of 80–150 K using non-

flammable components. Dobak et al. [11] obtained the patent for some non-flammable mixtures used in a miniature mixed gas refrigeration system for the temperature range of 110–190 K. Keppler et al. [12] have compared the performance of optimized hydrocarbon gas mixtures with that of optimized non-flammable hydrofluorocarbon mixtures. Luo et al. [13] have reported a no load temperature of around 50 K using auto cascade mixed refrigerant (AMR) cycle. Wang et al. [14] have reported the lowest temperature of 65.3 K using auto cascade system. They have used two liquid–vapor phase separators. However to the best of our knowledge, none of the researchers have reported no load temperatures lower than 70 K using single stage MR J–T cooler. Also, very few have used pressure–enthalpy (P–h) and temperature–enthalpy (T–h) charts for the lowest temperature prediction after throttling in these machines.

The authors report experimental results on single stage MR J–T system developed and tested successfully in the laboratory, for various compositions of mixtures. These mixtures are of different types, viz. the mixtures with higher percentage (greater than 50%) of flammable components, mixtures with all non-flammable components and the mixtures with minimum percentage of (less than 10%) flammable components. The authors report a record lowest temperature of 65 K achieved using gas mixture with more flammable components, while a temperature below 100 K is achieved with the gas mixture having minimum flammable components. The authors present the performance of the MR J–T cooler in terms of cool down time and cooling loads for various mixtures used in experimentation. A comparison between predictions and

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experimentally obtained temperature after throttling is presented for two different mixtures.

2. Thermodynamics of mixtures

The components used to form the refrigerant mixtures to be used for cryogenic applications are from three groups. Group-I has low boiling components (commonly referred as cryogens). Group-II contains light hydrocarbons and Group-III has chlorofluorocarbons. Table 1 gives the list of the components in different groups giving more stress on the gas components used in our experimentation. They are arranged in increasing order of normal boiling point (NBP) respectively in each group. Critical thermodynamic properties and latent heat of vaporization at one atmospheric pressure are also mentioned. The components, having NBP lower than 120 K, are referred to as low boiling components. From Group-II, methane can be referred to as low boiling component. Some other possible components are reported in the literature [16]. The main thermodynamic properties emphasized while selecting components are

(1) Normal boiling point (NBP).
(2) Specific refrigerating effect.
(3) Freezing temperature.
(4) Solubility of components at lowest working temperature.

The importance of the above properties for the mixture is described in brief. Normal boiling point of the lowest boiling component determines the possible lowest temperature that can be obtained with a given mixture. However, the lowest temperature that can be achieved from the mixture is determined by the saturation temperature corresponding to partial pressure of the lowest boiling component after expansion. Specific refrigerating effect, particularly of high boiling components, enhances the cooling capacity of the mixture and hence that of the system. Freezing temperature and solubility of the high boiling components in liquid nitrogen or in the low boiling components limit the lowest temperature that can be obtained from the mixture. To reach temperature of about 80 K, one needs to use at least one component from Group-I. To increase the cooling capacity and to operate MR J–T system within moderate pressure limits, one needs to select components from Group-II with appropriate composition. Group-III contains hydrofluorocarbon gases which are non-flammable components having less solubility in liquid nitrogen at low tem-

peratures [16]. As a result, their use in mixture reduces the performance of the cooler in terms of lowest temperature achieved and the cooling capacity. It should be noted that all the components from Group-II and Group-III have their freezing temperature above the NBP of nitrogen (N_2).

3. Experimental setup

Fig. 1 gives the experimental setup to evaluate MR J–T cooler performance with various mixtures. The closed cycle MR J–T cooler works analogous to standard vapor compression cycle used for commercial air conditioning systems. Oil lubricated rolling piston type rotary compressor (displacement 22 cc/rev) and air-cooled finned tubes after-cooler are used. Highly efficient three stage oil separators are placed after the after-cooler. First two oil separators are of sintered mesh type and third one is of coalescence type which can separate the oil up to 0.01 ppm. A recuperative counter flow tubes in tube type heat exchanger located after the oil separation section consists seven small tubes (I.D. = 2 mm, O.D. = 3 mm) inserted in one tube (I.D. = 10.7 mm, O.D. = 12.5 mm). The oil separators and heat exchanger are two additional subcomponents in MR J–T cycle as compared to standard vapor compression cycle. Capillary tubes are used as a throttling device. A small box type cold end heat exchanger is used as evaporator. An electric heater of 40 W capacity is mounted on the cold end to simulate cooling load. The cycle has five main processes:

(1) Adiabatic or polytropic compression.
(2) Heat rejection to coolant, either air or water.
(3) Cooling of high pressure stream by returning low pressure stream.
(4) Isenthalpic expansion (throttling) in throttle valve or capillary tube.
(5) Heat absorption at the cold tip/evaporator.

The compressor compresses the gas polytropically. The compressed fluid rejects heat to surrounding cooling medium which may be either air or water, in the after-cooler. After separating the oil in oil separators, the high pressure stream enters the heat exchanger. The high pressure supply stream is cooled by the low pressure return line in the heat exchanger. Further throttling of the fluid results in the low pressure, low temperature stream which, in turn, produces cooling effect at the cold end and then returns, through heat exchanger, to the compressor completing the

Table 1
List of the refrigerants used in present work[a].

Sr. no.	Name of refrigerant	T_{NBP} (K)	T_{FREEZ} (K)	T_C (K)	P_C (kPa)	$V_C \times 10^3$ (m^3/kg)	Q_{latent} (kJ/kg)
Group-I							
1	Helium (R704) He	4.25	Nil	5.25	228.8	14.43	20.75
2	Neon (R720) Ne	27.05	24.55	44.45	3397	2.07	
3	Nitrogen (R728) N_2	77.35	63.15	126.25	3396	3.179	198.84
Group-II							
4	Methane (R50) CH_4	111.65	90.95	190.65	4638	6.181	510.83
5	Ethane (R170) C_2H_6	184.35	90.15	305.35	4891	5.182	489.47
6	Propane (R290) C_3H_8	230.25	85.45	369.85	4248	4.53	425.43
7	Iso-butane (R600a) iC_4H_{10}	261.42	113.15	408.15	3645	4.526	366.69
Group-III							
8	Trifluromethane (R23) CHF_3	191.05	118.15	298.75	4833	1.942	238.68
9	Pentafluoroethane (R125) C_2HF_5	224.58	170	339.45	3630.6	–	163.38
10	R404A (R125 + R143a + R134a) (44/52/4)	226.67	–	345.65	3735	1.74	199.73
11	Chlorodifluromethane (R22) $CHClF_2$	232.93	113.15	369.15	4974	1.904	233.75
12	Tetrafluoroethane (R134a) CF_3CH_2F	246.99	176.55	374.25	4067	1.81	216.97
13	Difluoroethane(R152a) CHF_2CH_3	248.15	156.15	386.65	4492	2.741	329.91

T_{NBP} = normal boiling point, T_{FREEZ} = freezing temperature, T_C = critical temperature, P_C = critical pressure, V_C = critical volume, Q_{latent} = latent heat of vaporization.
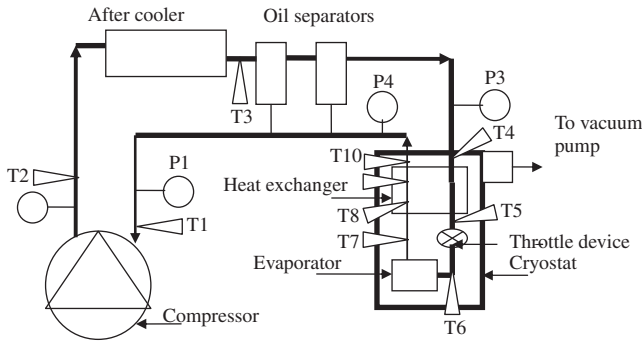[a] The properties of the refrigerants are from ASHRAE [15].

Fig. 1. Experimental setup with instrumentation.

cycle. It also shows instrumentation required to carry out pressure and temperature measurements at different locations. Locations P1–P4 represent the pressure gauges while T1–T10 indicate the temperature sensors. PT-100 temperature sensors are used to measure temperatures at suction of compressor, discharge from compressor, inlet and outlet temperatures for heat exchanger streams, and at cold tip inlet. Two additional temperature sensors are placed on the return line of counter flow heat exchanger. The electrical power consumption of the compressor is measured with digital Watt meter (not shown in figure).

For a given set of major components, i.e. the compressor and the heat exchanger, the parameters that affect the performance of MR J–T coolers are charging pressure, capillary tube (throttling device) dimensions and composition of the mixture. While the charging pressures are determined by the compressor selection, the capillary tube dimensions could be worked out by trial and error

method. The effect of charging pressure and geometry of capillary on the performance of MR J–T cooler is explained elsewhere [17]. For a given set of these parameters, the composition of mixture is the major factor which determines the power input, lowest achievable temperature and the cooling capacity obtained from the cooler. Experiments are carried out with mixtures formed with nitrogen–hydrocarbons (N₂–HC), neon–nitrogen–hydrocarbons (Ne–N₂–HC), nitrogen–hydrocarbons–hydroflurocarbons (N₂–HC–HFC), and nitrogen–hydroflurocarbons (N₂–HFC) components.

Mixtures are prepared by charging the required components of gases in the system according to their partial pressures based on the mixture composition and total charging pressures. The mixture compositions are checked using gas chromatograph and an average deviation of about 5% is observed in composition. In addition, some experiments are carried out using a third party calibrated N₂–HC mixture for confirming the results.

4. Results and discussion

4.1. P–h and T–h diagram of Neon-11 mixture

Based on the selection criteria discussed above, the components are selected from three groups and the thermodynamic properties of mixtures are studied for different compositions. A systematic study is carried out starting with binary composition with subsequent addition of different components in the mixture. The properties are calculated using software aspenONE version 2004.1 [18] using Peng–Robinson equation of state [19]. P–h and T–h diagrams are plotted for different mixtures. Fig. 2 shows the P–h diagram for Neon-11 mixture with experimental data points 1–6. Description of points on P–h chart is mentioned below Fig. 2. The mixture
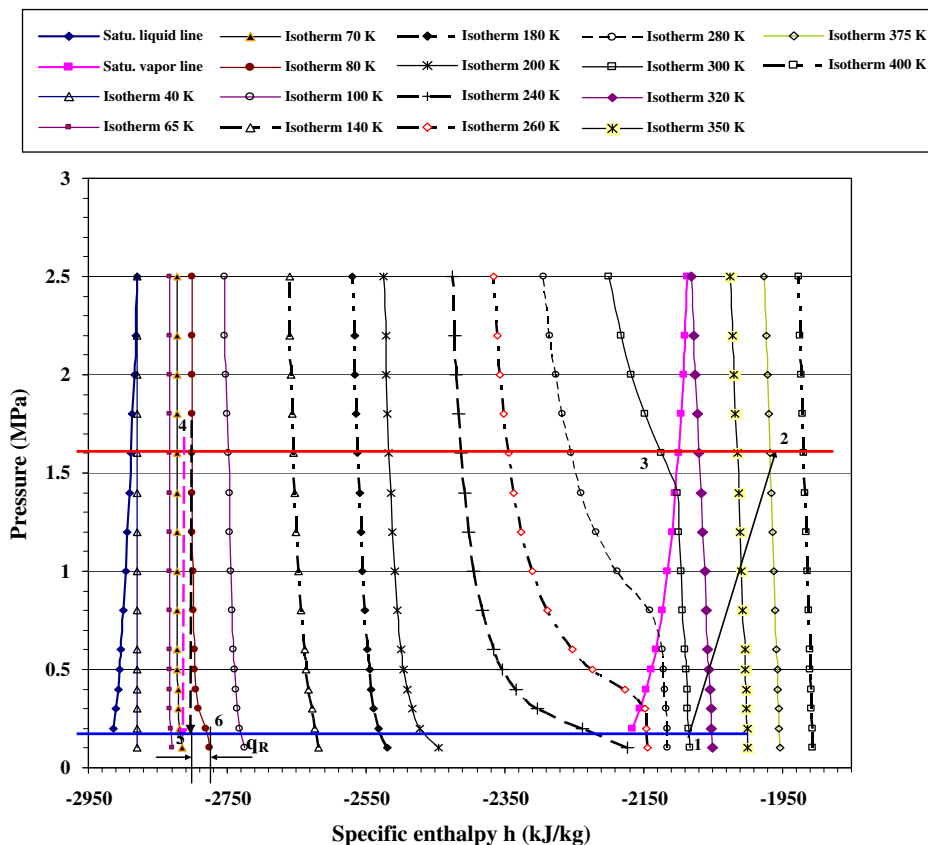

Fig. 2. P–h diagram for Neon-11 mixture. 1 – Compressor suction, 2 – compressor discharge, 3 – outlet of after-cooler or high pressure stream inlet to heat exchanger, 4 – high pressure stream before throttling, 5 – cold tip inlet, 6 – cold tip outlet.

consists of six components namely neon (Ne), nitrogen (N$_2$), methane (CH$_4$), ethane (C$_2$H$_6$), propane (C$_3$H$_8$) and iso-butane (iC$_4$H$_{10}$). The overall percentage mol of low boiling components (Ne, N$_2$, CH$_4$) is about 53% while percentage mol of flammable components (CH$_4$, C$_2$H$_6$, C$_3$H$_8$ iC$_4$H$_{10}$) is about 70%. Different isothermal lines are also shown in Fig. 2. The figure shows an actual cycle with the operating pressure limits of 1.6 MPa and 0.18 MPa. (The pressure and temperature values are taken from actual experimental trial conducted.) Dew point temperature for the mixture Neon-11 at 1.6 MPa is about 303 K which is above the normal ambient temperature (300 K). This indicates that some of the high boiling components condense before entering the heat exchanger. Note that the point 3 indicating the inlet of high pressure stream to heat exchanger is inside the liquid–vapor region and close to 300 K. The high pressure stream entering the heat exchanger (point 3) is in two phase condition. Due to this, heat transfer coefficient is high and thus improves the performance of heat exchange process and hence, the cooler performance. Specific enthalpy difference between the inlet to cold tip (point 5) and outlet of cold tip (point 6) represents the specific cooling effect (q_R) that can be obtained for the given mixture within the given working parameters. It can be observed from the Fig. 2 that the major part of the cycle is in the liquid–vapor zone of the P–h diagram.

Fig. 3 shows T–h diagram for mixture Neon-11 with the high pressure limit of 1.6 MPa and lower pressure limit of 0.2 MPa. It indicates the lowest possible temperature that could be attained using the mixture, for the given pressure limits. The lowest temperature that can be achieved theoretically with this mixture is about 58 K for the given pressure limits, as shown in Fig. 3 with a dash.

4.2. Performance curves for Neon-11 mixture

Fig. 4 shows the cool down curves for the Neon-11 mixture. The pressure reaches the maximum value in the beginning to 2.3 MPa due to higher average temperatures of the working space while as the temperature gets lowered the pressure gets reduced down to 1.6 MPa due to condensation of refrigerants as well as lowering of average temperature of the working space. The compressor suction pressure stabilizes at about 0.18 MPa. It is observed that for the mixtures containing neon or helium, the initial pressure ratio is higher than 10 and the same stabilizes at about 8–9. It is higher than that for the N$_2$–HC mixtures by 40–60% depending on the composition of the mixture. The temperature below 100 K is reached in one hour while the lowest temperature of 65 K is achieved in around three and half hours. The experimentally obtained lowest temperature is higher than the predicted lowest temperature of 58 K. This is due to the fact that effects of pressure
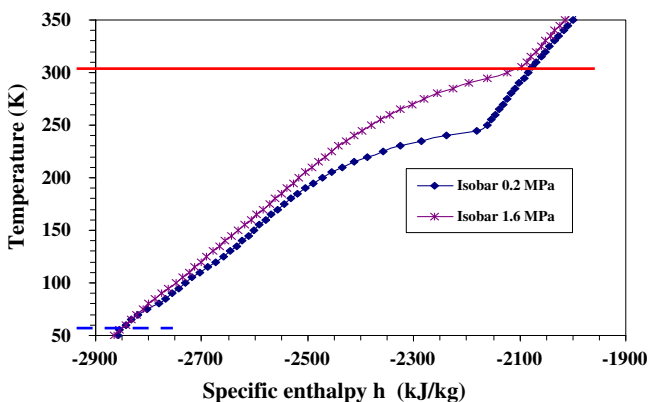
drop, external heat leakage (estimated heat gain 2.8 W at 80 K) and heat exchanger effectiveness, etc. are not considered in the theoretical predictions.

It can be seen that the slope of the temperature curves changes as the different components gradually condense during the cool down. For example, the slope of the temperature curve before throttling changes at about 120 K. At this temperature and pressure of 2.0 MPa, all the high boiling components (iC$_4$H$_{10}$, C$_3$H$_8$, C$_2$H$_6$) are in liquid state. The percentage of liquid methane is about 93.5% of its total mass while percentage of liquid nitrogen is about 32% of its total mass. As the system moves towards the steady state condition, the pressure and the temperature of the high and the low pressure streams stabilize and hence the percentage of liquid and vapor state of each component also stabilizes at a certain fixed proportion.

Fig. 5 shows the cooling load and power consumption of compressor for the same system. A cooling capacity of 6.1 W is obtained at 80 K and of about 21 W is obtained at 100 K. It is clear from the graph that cooling load varies linearly with respect to cold tip outlet temperature. The compressor power consumption increases gradually with increments in the load. A power consumption of 1031 W is noted for the cooling load of 28.73 W at 108 K; while power consumption for cooling load of 6.1 W at 80 K is found to be 868 W. Cooling capacity and the compressor power input
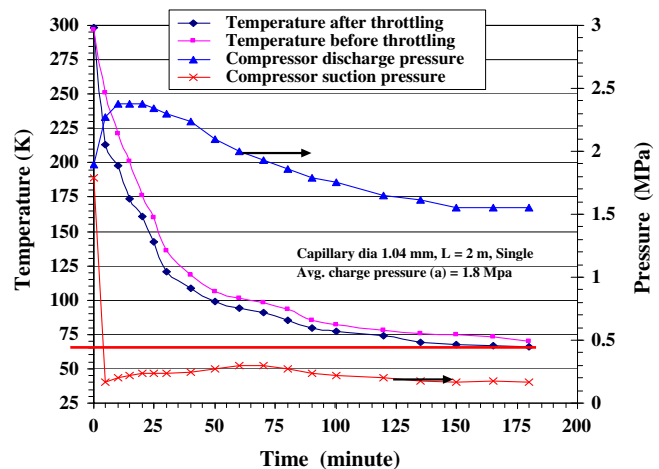

Fig. 4. Cool down performance for Neon-11 mixture.

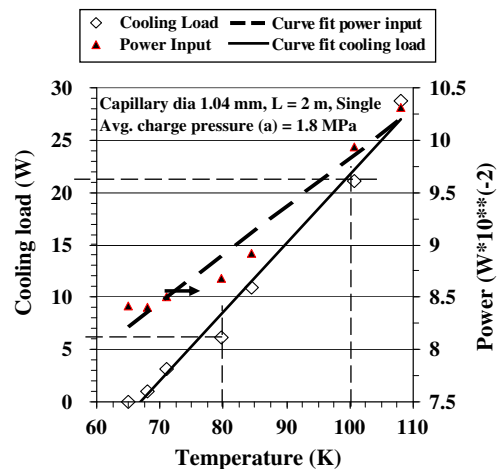**Fig. 3.** T–h diagram for Neon-11 mixture.

Fig. 5. Cooling load performance of Neon-11 mixture.

show linear trend in the given temperature range, as can be seen from the Fig. 5. Coefficient of performance (COP) is defined as a ratio of cooling capacity at cold tip to power input to the compressor. Fig. 6 shows the variation of COP with respect to cold tip outlet temperature. The COP obtained at 80 K is 0.00708. While the maximum COP obtained is 0.0278 at 108 K.

The performance of the system can be improved by optimizing the mixture, optimizing the heat exchanger and the system as a whole. The present mixture, Neon-11, is selected so as to achieve the lowest temperature. For COP optimization, a different optimized mixture composition is required.

4.3. Prediction of temperature after throttling

Prediction of temperature after throttling from a given high pressure and temperature condition to lower pressure is an important design aspect of the cooling system. This temperature can be predicted within a close accuracy using T–h charts of mixture. Fig. 7 shows the comparison of predicted and experimental temperatures after throttling for the same mixture Neon-11. The temper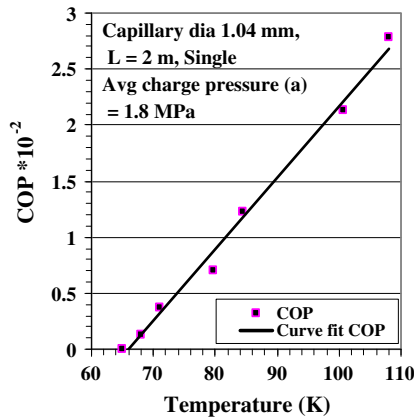ature data, in terms of temperature before and after throttling, is collected during the transient period of cool down where the high pressure decreases from 2.3 MPa to 1.6 MPa while the low pressure increases from 0.16 MPa to 0.23 MPa. As a result, an average value of high and low pressures, 1.8 MPa and 0.2 MPa, are considered in this case. These isobars are plotted on T–h diagram. Isenthalpic expansion lines drawn from high pressure isobar and temperature before the throttling (recorded as T_5 in Fig. 3) on low pressure isobar, when projected on temperature axis of T–h diagram, gives predicted value of temperature after throttling within given averaged pressure limits. Due to this an average error of 1.81 K is observed between predictions and experimentally obtained temperature after throttling.
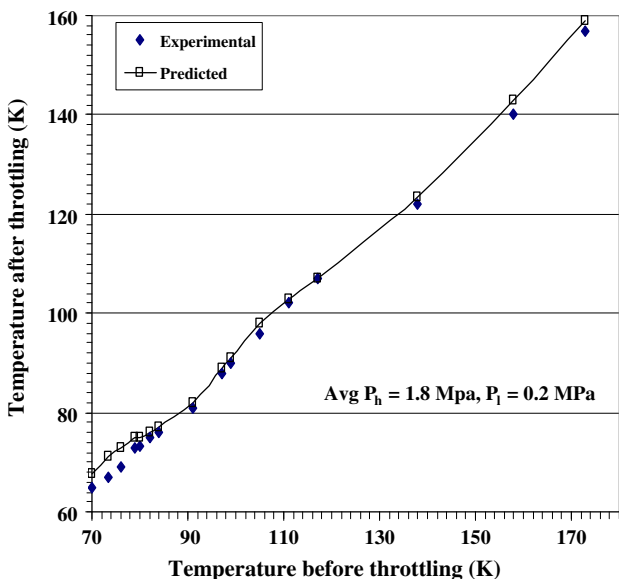
In order to confirm the validity of the above results, same comparison is carried out for other mixture with the high and low pressure conditions of 1.6 MPa and 0.4 MPa respectively. Fig. 8 shows the comparison of prediction and experimentally obtained temperatures after throttling for the quaternary N_2–HC mixture-5. The average difference between the prediction and experimentally obtained temperature after throttling for the same is found to be 1.24 K. Close average deviations in both the cases discussed above confirm the usefulness of the T–h diagram to predict the temperature after throttling with close accuracy in MR J–T coolers.

4.4. Performance curves for non-flammable (NF) and semi-flammable (SF) mixtures

Experimental trials are conducted using NF mixtures consisting of gases such as R22, R125, R134a, R404A, R23, N_2 and Ne. Fig. 9 shows the cool down characteristics for the mixture consisting of R134a, R404A, R23 and N_2 (NFmixture-4) charged at average pressure of about 1.7 MPa. Broadly it has 43% mole of low boiling component (N_2). The lowest temperature recorded with this mixture is about 122 K with the maximum compressor discharge pressure of about 2.27 MPa stabilizing down to 1.7 MPa. The suction pressure variation is very small and it remains stable about 0.16 MPa.

In order to improve the performance, a flammable gas, methane is added up to 10% to the above mentioned NFmixture-4. This mixture is referred to as a semi-flammable mixture (SFmixture-5) and is used for further experimentation. It contains about 48% low
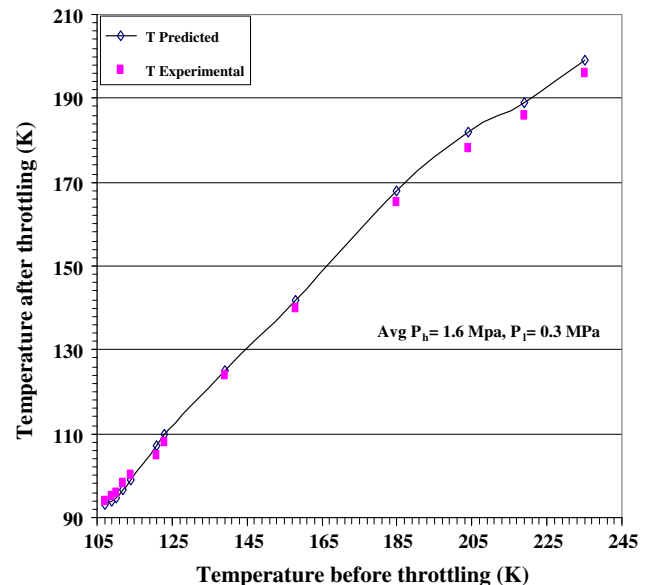

Fig. 6. Variation of COP of Neon-11 mixture.

Fig. 7. Comparison of prediction and experimental temperatures after throttling for Neon-11 mixture.

Fig. 8. Comparison of prediction and experimental temperatures after throttling for N_2–HC mixture.
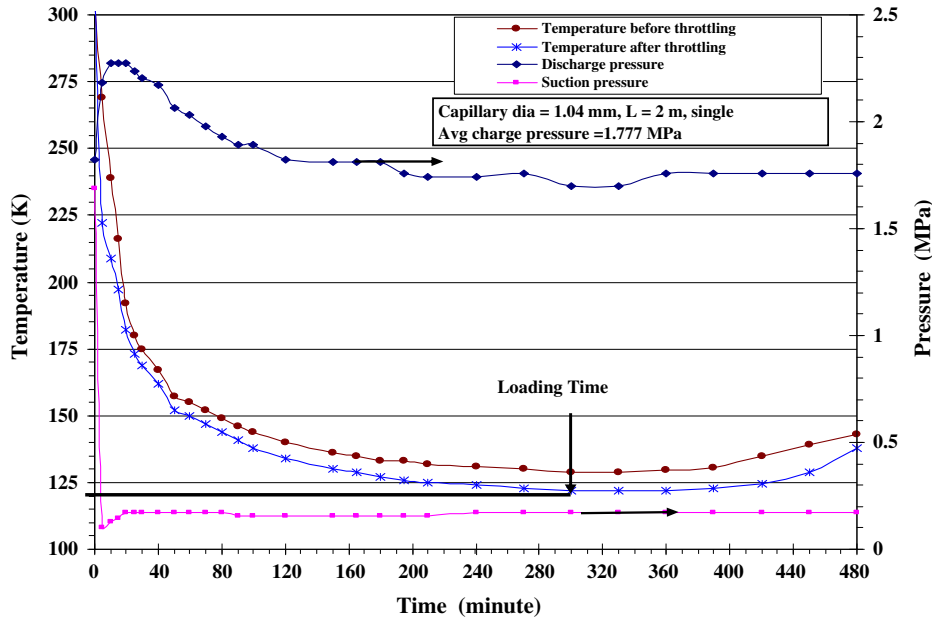
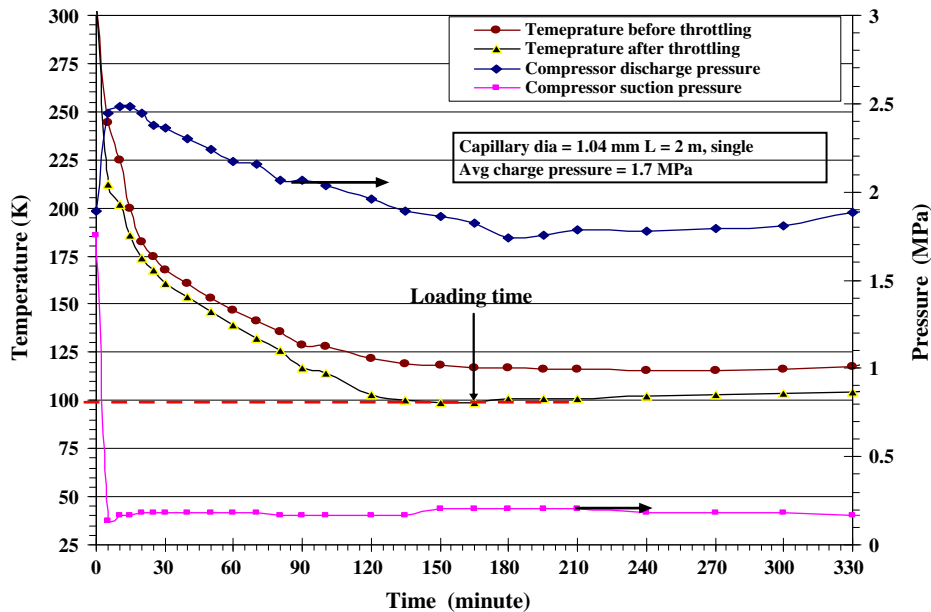**Fig. 9.** Cool down performance for NFmixture – 4.

Fig. 10. Cool down performance for SFmixture – 5.

boiling components (N_2 and CH_4). Fig. 10 shows cool down curves for SFmixture-5 for the charged pressure of about 1.7 MPa. The lowest temperature recorded with this mixture is 99 K. It is obvious that the lower temperature achieved in this case is due to presence of the small percentage of methane. The maximum compressor discharge pressure reaches to 2.3 MPa and stabilizes at about 1.9 MPa. The suction pressure variation is very small and it remains stable about 0.18 MPa.

Fig. 11 represents the cooling load characteristic for SFmixture-5 and NFmixture-4. Using SFmixture-5 a cooling capacity of 4 W is obtained at 120 K. While using NFmixture-4, the same cooling capacity is obtained at temperature of 143 K. The power input to compressor is of about 873 W for SFmixture-5 and 792 W for

NFmixture-4 over the given cooling loads range. The nature of the cooling load with cold tip outlet temperature is a polyline for SFmixture-5 while it is linear for NFmixture-4 over the given cold tip temperature range. The initial flatter curve obtained for SFmixture-5 is due to sensible cooling effect contributed by methane and nitrogen. Due to this, there is a sudden rise in the cold tip temperature from 99 K to 111 K for a very small load of 0.5 W. Further cooling effect is obtained mainly due to latent heat contribution of high boiling hydrochloroflurocarbon components.

Fig. 12 gives the variation of COP against cold tip outlet temperature for SFmixture-5 and NFmixture-4. The trend lines for both the mixtures are similar to those obtained in the respective cooling load characteristics. The maximum COP obtained for SFmixture-5
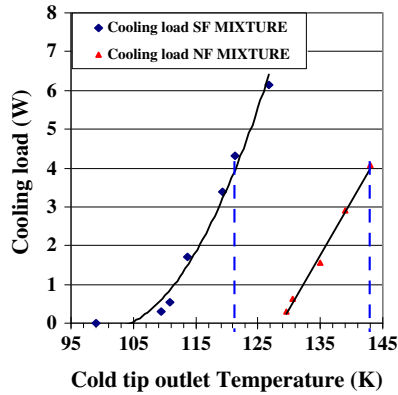
Fig. 11. Cooling load performance of SFmixture – 5 and NFmixture – 4.
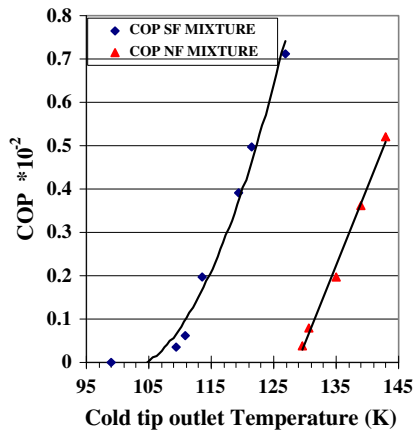

Fig. 12. Variation of COP of SFmixture – 5 and NFmixture – 4.

is of about 0.007 at 127 K as against 0.005 COP is obtained for NFmixture-4 at 143 K.

5. Conclusions

- A single stage mixed refrigerant Joule–Thomson cryocooler has been successfully developed and tested in our laboratory and a record low temperature of 65 K is achieved using Neon-11 mixture. Cooling capacity of about 6 W at 80 K is obtained with power input of 868 W.
- The utility of P–h and T–h diagrams for different types of mixtures is justified in the present work.

- The experiments show that with non-flammable mixtures, the cooling capacity gets reduced while there is substantial increase in the lowest temperature achieved as compared with N_2–HC, Ne–N_2–HC mixtures. Non-flammable mixtures are more suitable for relatively high temperature applications.
- Use of mixtures containing minimum flammable components, such as SFmixture-5, could be one of the methods to improve the performance of cooler working in relatively high temperature range.

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