Thermodynamic optimization of mixed refrigerant Joule-Thomson systems constrained by heat transfer considerations

J F Hinze, S A Klein, G F Nellis
University of Wisconsin-Madison
1500 Engineering Drive, Madison, WI, USA
Email: jfhinze@wisc.edu

Abstract. Mixed refrigerant (MR) working fluids can significantly increase the cooling capacity of a Joule-Thomson (JT) cycle. The optimization of MRJT systems has been the subject of substantial research. However, most optimization techniques do not model the recuperator in sufficient detail. For example, the recuperator is usually assumed to have a heat transfer coefficient that does not vary with the mixture. Ongoing work at the University of Wisconsin-Madison has shown that the heat transfer coefficients for two-phase flow are approximately three times greater than for a single phase mixture when the mixture quality is between 15% and 85%. As a result, a system that optimizes a MR without also requiring that the flow be in this quality range may require an extremely large recuperator or not achieve the performance predicted by the model. To ensure optimal performance of the JT cycle, the MR should be selected such that it is entirely two-phase within the recuperator. To determine the optimal MR composition, a parametric study was conducted assuming a thermodynamically ideal cycle. The results of the parametric study are graphically presented on a contour plot in the parameter space consisting of the extremes of the qualities that exist within the recuperator. The contours show constant values of the normalized refrigeration power. This ‘map’ shows the effect of MR composition on the cycle performance and it can be used to select the MR that provides a high cooling load while also constraining the recuperator to be two phase. The predicted best MR composition can be used as a starting point for experimentally determining the best MR.

1. Introduction
Joule-Thomson (JT) cycles have many different uses as they provide a compact, simple, and low vibration cooling load. They have been used for cryosurgery as well as for cooling low temperature sensors. However their operation makes them inherently less efficient than Stirling cycle coolers [1]. In order to compete with other cycles, the cooling power of JT cycles needs to be increased while reducing the overall size of the device. These improvements will allow JT cycles to be used for a wider range of applications.

Research has shown that Mixed Refrigerant Joule-Thomson (MRJT) cycles have the ability to dramatically increase cooling load while decreasing the operating pressures when compared to a pure refrigerant JT cycle [2]. However, this increase in performance depends greatly on the selection of Mixed Refrigerant (MR) used in the cycle. An improperly selected mixture can result in reduced cooling load or even system failure due to solidification in the expansion valve. Mixture selection is usually done by first modeling the cycle and determining the optimal mixture composition followed by experimentally verifying the results [1, 3]. Since existing models do not accurately model the heat transfer occurring in the recuperator, this process can require extensive experimental testing.

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd
The main deficiency of previous MRJT cycle models (e.g., [1, 2, 3]) has been that they do not explicitly consider the enhancement of the heat transfer coefficient of the MR that occurs in the two phase region. Work done at the University of Wisconsin has shown that the heat transfer coefficient for mixed refrigerants in the two phase region can be as much as three times greater than the heat transfer coefficient in the sub-cooled or super-heated regions [4]. Figure 1 shows the heat transfer coefficient as a function of quality for a representative mixed refrigerant. The heat transfer coefficient in the subcooled and super-heated regions are indicated by arrows at quality of zero and one.

Figure 1. Heat transfer coefficient (htc) Vs average quality (x_{avg}) for hydrocarbon mixed refrigerant. From [4].

A higher heat transfer coefficient allows the cooling system to provide a larger cooling load for a fixed size of the recuperator. Therefore, the optimal mixture is one that will provide the highest cooling load while also having a high heat transfer coefficient (i.e., a two-phase condition) throughout the recuperator. Figure 1 shows that the heat transfer coefficient is high when the quality is in the range of 15% to 85% and this trend was observed for every mixture and condition that was tested. This means that the optimal mixture is one that provides the highest cooling load while also remaining in this quality range within the recuperator. The goal of this research is to understand the difference between the thermodynamically optimized mixture, the mixture that provides the theoretical maximum possible cooling load independent of heat transfer rate considerations, and the heat transfer constrained optimized mixture, which is the mixture that provides the highest cooling load while remaining between 15% and 85% quality in the recuperator. By understanding the effect that the heat transfer coefficient enhancement has on mixture selection, the model should more accurately be able determine the optimal mixture, resulting in less experimental testing.

2. MRJT cycle model
A model was created that can calculate the cycle cooling power for a given set of parameters using Engineering Equation Solver (EES). REFPROP is used to calculate the states of the MR in the cycle. It was found that REFPROP had a lower error rate when using mixtures of hydrocarbon refrigerants compared to synthetic refrigerants. Therefore, this research initially focused on hydrocarbon mixtures.
The states of the hydrocarbon mixtures are calculated according to the GREG 2008 equation of state which is valid for the range of mixtures covered [5].

The model divides the MRJT cycle into parts; the first part consists only of the compressor and the second part consists of only the cold head of the cycle as seen in Figure 2. A third part provides heat exchange between the pure refrigerant in a higher temperature conventional cycle and the MR cycle, if it is employed for a pre-cooled system.

![Figure 2. MRJT total system diagram with sub-models identified.](image)

The heat exchanger for the reheat of the MR back to room temperature is not explicitly modeled and rather it is assumed that it is sufficiently large to provide the needed heating load. Therefore, the MR is assumed to be at room temperature at the entrance to the compressor. Similarly it is assumed that the after cooler and pre-cooling heat exchangers are sufficiently large to cool the MR to room temperature and the pre-cooler temperature, respectively. These simplifications allow a pre-cooled cycle to be modeled simply by specifying a lower supply temperature to the cold head model. This idealized model also assumes that all of the pressure drop through the cycle occurs at the expansion valve, with no pressure drop occurring in the heat exchangers.

The compressor is modeled using a polytropic compression process, based on work done by Jähnig [6]. The compressor model input parameters are the suction pressure, discharge pressure, and mixture composition and the model returns the volumetric efficiency of the compressor according to equation (1).

\[ \eta_v = 1 + \frac{C}{P_{\text{discharge}}} \left( \frac{P_{\text{discharge}}}{P_{\text{suction}}} \right)^{\frac{1}{k}} \]  

(1)
Where $C$ is the clearance volume fraction, which is assumed to be 0.025 \cite{6}. The parameter $k$ is the specific heat ratio which is calculated at the suction side of the compressor using REFPROP. The mass flow rate of refrigerant through the cycle can then be calculated using equation (2).

$$m = \eta\, \rho_{\text{suction}}\, \dot{V}_{\text{displace}}$$  \hspace{1cm} (2)

Where $\rho_{\text{suction}}$ is the density at the suction of the compressor, and $\dot{V}_{\text{displace}}$ is the displacement rate of the compressor.

The cold head model includes the recuperator, expansion valve, and load heat exchanger as seen in Figure 2. The temperatures at states 1 and 4 are inputs to the cold head model, and the mass flow rate, suction pressure, and discharge pressure are known from the compressor model. The pressures at states 1 and 2 are equal to the discharge pressure, and at states 3, 4, and 5 are equal to the suction pressure. The temperature at state 1 is the supply temperature and can be changed to model either a room temperature cycle or a pre-cooled cycle.

The recuperator is modeled by specifying the pinch point temperature difference, which is the smallest temperature difference between the high pressure and low pressure streams in the recuperator. The smaller the pinch point temperature difference, the more heat is exchanged in the recuperator. A pinch point temperature difference of zero corresponds to a perfect recuperator that has the highest possible performance. This model specifies the pinch point temperature difference to be 0 K which is consistent with a thermodynamically optimal system; the predicted performance is a result of only the thermodynamics of the MR. By expressing the results in the coordinate space of recuperator quality extremes we can examine how easy or hard it will be to approach this ideal limit. A more realistic simulation will adjust the pinch point temperature difference based on the heat transfer coefficients in the recuperator. However, this process becomes computationally very difficult, particularly in the context of a thorough optimization process which requires many simulations. We can assume based on measurements that when the MR in the recuperator is in the range of 15\% and 85\% the cycle will operate with a small pinch point temperature difference. To solve for the pinch point temperature difference, the heat exchanger is broken into sub-sections, each with equal heat transfer, as described in \cite{7}. The enthalpy at state 2 is adjusted iteratively until the specified pinch point temperature difference is achieved.

From states 2 to 3 the MR undergoes isenthalpic expansion so the enthalpy and pressure at state 3 are known. The cooling load normalized against the compressor displacement rate is calculated according to:

$$Q_{\text{load, norm}} = \frac{m(h_s - h_3)}{\dot{V}_{\text{disp}}}$$  \hspace{1cm} (3)

To determine the optimal MR, a parametric study was run using the model defined above. A pre-cooled cycle was modeled using a load temperature of 150 K and a supply temperature of 240 K; these conditions are consistent with a commercial cryosurgical probe cycle. The refrigerants chosen were methane, ethane, and butane which have normal boiling points of 111.7 K, 184.6 K, and 272.7 K respectively \cite{8,9,10}. It is important that the components used in the mixture have boiling points that span the range of temperatures in the recuperator so that the MR exhibits the appropriate temperature glide. If the components are improperly chosen, the MR will not remain two phase and the performance will suffer thermodynamically as well as from a heat transfer perspective. The suction pressure is held constant at 150 kPa in this study. This optimization looked at the effect of changing the composition and the discharge pressure of the cycle simultaneously. The mole fraction parameter space contained every possible combination of mole fractions, meaning each mole fraction each component varied from zero to one. The parameter space for the discharge pressure ran from 1400 kPa to 3700 kPa in 100 kPa intervals, this was found to cover all results of interest. For each run of the parametric study, the normalized cooling load (defined as the cooling load per unit of compressor
displacement) was recorded, as well as the limits of quality in the recuperator (i.e., the highest and lowest values). Note that negative qualities correspond to subcooled conditions and qualities greater than 1 correspond to superheated conditions.

3. Model predictions

A graphical representation of the results was created to gain a better understanding of the results of the parametric study. The independent variables were the limits of quality in the recuperator. The x axis is the lowest quality in the recuperator which exists at the high pressure exit of the recuperator, and the y axis is the highest quality in the cycle which exists at the low pressure exit of the recuperator. Contours of constant normalized cooling power (divided by the maximum value observed at any condition) are plotted in this parameter space. Lines showing the location of the different quality constraints are shown; these lines indicate the region where the cycle would operate with enhanced heat transfer coefficients. The plot shows the relationship between the quality in the recuperator and thermodynamic cooling power of the cycle. For any combination of quality limits within the area defined by the contours, there is a MR that will provide a cooling power corresponding to its location on the contours. By plotting our results in this fashion, a ‘map’ of the cycle is created showing the relationship between quality limits and the normalized cooling load. The ‘map’ created for the pre-cooled cycle is shown in Figure 3.

![Figure 3](image.png)

**Figure 3.** Contour plot of cooling power for hydrocarbon three component mixture operating between 240K and 150K with a suction pressure of 150 kPa.

There are several locations of interest that are identified on the plot. The first is the thermodynamic maximum point, represented by a white dot. This point corresponds to the MR that provides the highest normalized cooling power. There are three other points that represent the cooling power while taking into consideration the quality limits in the recuperator. The first point is the maximum cooling load when the quality in the recuperator is between 0 to 100 percent, the second when between 10 and 90 percent and lastly when between 15 and 85 percent. The cooling load, pressure, and composition at these points are identified.
Figure 3 shows that the thermodynamic maximum cooling load occurs when the MR in the recuperator exists as a slightly sub-cooled liquid and a slightly super-heated vapor at the high pressure exit and low pressure inlet, respectively. Clearly, this point is well outside of the constraint that the quality in the recuperator must be between 15 and 85 percent to ensure good heat transfer. To meet this constraint, the theoretical cooling load must be reduced significantly to about 55 percent of the thermodynamic maximum possible value. If the constraint is relaxed allow the quality in recuperator to be within 10 and 90 percent then the cooling load is increased to about 62 percent of the maximum possible value. If the constraint is further relaxed so that the MR must only be two phase in the recuperator, i.e., between 0 and 100 percent quality, then the cooling load is increased to almost 80 percent of the maximum cooling load. This large penalty in cooling load is necessary in order to have good heat transfer coefficients in the recuperator and it complicates the determination of the truly optimal mixture composition.

Identification of the best mixture composition is an optimization problem where moving to better heat transfer regions causes lower thermodynamic cooling. As a result, there must be an optimal point where the loss of cooling load is offset by the increase in performance as a result of the enhanced heat transfer coefficient. Figure 3 can be used to identify which MR might fit the description of the optimal mixture, good heat transfer coefficients with a high cooling load.

Figure 3 also shows a white line connecting the thermodynamic maximum point to the points of maximum cooling power for a given quality constraint. This will be referred to as the design line and it is along this line that the optimal mixture must reside. This line represents the highest cooling load possible for a given constraint on quality. In order to determine the best mixture along this design line, the points need to be studied in more detail. In the initial analysis it was assumed that the system would not perform as designed if the recuperator was not operating in the good heat transfer region of quality. To test this theory, the cycle that provided the maximum thermodynamic cooling power and the cycle that provided the highest cooling load when constrained to be within 15 and 85 percent were both plotted on a P-h diagram for their respective mixtures, as seen in Figure 4 and Figure 5.

![Figure 4](image-url)  
*Figure 4. Pressure-enthalpy diagram of thermodynamic mixture identified in Figure 3.*
Figure 5. Pressure-enthalpy diagram of 15 to 85 percent quality constrained mixture identified in Figure 3.

It is clear that, for the constrained case in Figure 5, all of the MR in the recuperator is between 15 to 85 percent quality where the heat transfer coefficients are high. However, the P-h diagram for the thermodynamic maximum mixture in Figure 4 shows that about two thirds of the MR in the recuperator is in the good heat transfer region. Meaning that the heat transfer coefficient degradation will only affect a third of the recuperator. It is predicted that the MR that provides the best real world cooling performance will have a high thermodynamic cooling load while operating with most of the recuperator in the good heat transfer region. In order to verify this, the four points along the design line should be tested experimentally.

4. Future work and conclusions
The optimal mixture predicted by the model needs to be experimentally tested to ensure the actual optimal mixture has been identified. An experimental test facility at the University of Wisconsin will be used for testing. The facility offers good control of the cycle parameters and precise measurement of temperatures, pressures, and cooling load of the cycle. The four mixtures identified along the design line will be tested to determine the effect the heat transfer coefficient and thermodynamic cooling load have on the actual cooling load of the cycle. The composition of the charged mixture will be verified using a gas chromatograph. Using the controls of the test facility the parameters from the model will be matched as closely as possible to the actual cycle operating parameters. The cooling load at steady state for each mixture will be recorded. The measured cooling loads should show that the mixtures operating in the enhanced heat transfer region have the highest cooling load. The cooling load from this optimal mixture can be compared to the cooling load of the original mixture at the same temperature. If the optimal mixture has a cooling load similar to the original mixture this model has proved it can provide a good starting mixture for experimental testing, since the original mixture was a result of extensive experimental testing. The results of the experimental tests can also be plotted in the same parameter space as Figure 3, by using the measured temperature, pressure, and cooling load data from the experimental test facility. The modified plot should show that the actual cooling load peak is
closer to the enhanced heat transfer region than the thermodynamic cooling load peak identified by the white dot in Figure 3. This would prove that considering the effect of the enhanced heat transfer region is important when choosing a mixture for a MRJT cycle, and this optimization technique is valid.

This design method has the potential to accurately predict the best refrigerant for any given MGJT cycle more accurately and quickly than other models such as [1, 3]. Additional parametric studies were conducted which investigated the effects of changing the type and number of components, the temperature span, the suction pressure, and the pinch point temperature difference. These results indicate that this optimization method applies regardless of the cycle parameters [11]. If this model is proven accurate by experimental testing, it has the potential to greatly reduce the time needed to determine the optimal mixture because it recognizes it is impossible to match the parameters from a complicated model to the real world conditions. Instead, using a simple model and a powerful plotting technique, it can determine a range of optimal mixtures very quickly while also considering the effects of heat transfer. Experimental testing time can also be reduced by identifying four mixtures along the design line which should have a composition closer to the optimal mixture than those found using other optimization techniques. This means fewer mixtures need to be tested, also saving time.

5. References

[1] Fredrickson K, Nellis G and Klein S 2006 A design method for mixed gas Joule-Thomson refrigeration cryosurgical probes *Int. J. of Refrigeration* **29**(5) 700-15
[2] Arkhipov V, Yakuba V, Lobko M, Yevdokimova O and Stears H 2002 Multicomponent gas mixtures for J-T cryocoolers *Cryocoolers* **10** 487-95
[3] Keppler F, Nellis G and Klein S 2004 Optimization of the composition of a gas mixture in a Joule-Thomson cycle *HVAC&R Research* **10**(2) 213-30
[4] Barraza R 2015 Thermal-fluid behavior of mixed refrigerants for cryogenic applications PhD [dissertation] Madison: Univ. of Wis. Madison Available from: UW Madison Library System
[5] Kunz O and Wagner W 2012 The GREG-2008 wide-range equation of state for natural gases and other mixtures: an expansion of GERG-2004 *J. of Chem. & Eng. Data* **57** 3032-91
[6] Jähnig D 1991 A semi-empirical method for modeling reciprocating compressors in residential refrigerators and freezers MS [thesis] Madison: Univ. of Wis. Madison Available from: UW Madison Library System
[7] Nellis G and Klein S 2009 *Heat Transfer* (New York, NY: Cambridge University Press)
[8] Setzmann U and Wagner W 1991 A new equation of state and tables of thermodynamic properties for methane covering the range from the melting line to 625K at pressures up to 1000 MPa *J. Phys. Chem. Ref. Data* **20**(6) 1061-151
[9] Buecker D and Wagner W 2006 A reference equation of state for the thermodynamic properties of ethane for temperatures from the melting line to 675 K and pressures up to 900 MPa *J. Phys. Chem. Ref. Data* **35**(1) 205-266
[10] Buecker D and Wagner W 2006 Reference equations of state for the thermodynamic properties of fluid phase n-butane and isobutane *J. Phys. Chem. Ref. Data* **35** (2) 929-1019
[11] Hinze J Thermodynamic optimization of mixed refrigerant Joule-Thomson cycle with heat transfer considerations MS [thesis] Madison: Univ. of Wis. Madison Available from: UW Madison Library System