

Sharif University of Technology Scientia Iranica Transactions B: Mechanical Engineering

nsactions B: Mechanical Engineerii www.scientiairanica.com



# Design of mixed refrigerant cycle for low temperature processes using a thermodynamic approach

## M. Mafi<sup>a</sup>, B. Ghorbani<sup>b,\*</sup>, M. Amidpour<sup>b</sup> and S.M. Mousavi Naynian<sup>b</sup>

a. Department of Mechanical Engineering, Imam Khomeini International University, Qazvin, P.O. Box 34149-16818, Iran.
b. Faculty of Mechanical Engineering, K.N.Toosi University of Technology, Tehran, P.O. Box 19395-1999, Iran.

Received 20 June 2012; accepted 11 March 2013

KEYWORDS Mixed refrigerant cycle; Low temperature processes; Systematic design; Optimal operating conditions; Optimal arrangement. **Abstract.** Minimizing the work consumed by a refrigeration system is an effective measure for reducing the cost of products in sub-ambient chemical processes, such as olefin plants. A recent advancement has been the introduction of mixed working fluids in refrigeration systems in place of pure working fluids. Due to the lack of a systematic design method for the Mixed Refrigerant Cycle (MRC), conventional approaches are largely trial-and-error and, therefore, operations can be under far from optimal conditions. In this paper, a novel method for systematic design of MRCs is presented, which combines the benefits of the thermodynamics approach and mathematical optimization. Based on the success of the proposed systematic method for the optimal selection of refrigerant composition and operating pressures, the method is extended to give optimal arrangement of the cycle components. The procedure is demonstrated using a case study of the design of MRC for a typical olefin plant.

© 2013 Sharif University of Technology. All rights reserved.

## 1. Introduction

For many years, cascade refrigeration cycles have been used to cool and liquefy feed streams in subambient processes, such as olefin (ethylene recovery) plants. Such cascade cycles have commonly included a plurality of individual refrigerants having decreasing atmospheric boiling points, each of which is circulated in a closed cycle to heat exchange with the feed streams. Unfortunately, the use of such individual refrigerants requires a very large number of separate heat exchangers, pumps, compressors and associated pipes and valves for the separate closed loops of each state. Even more importantly, the cooling curves of individual refrigerants do not closely match the continuous cooling curve of the feed stream, and this is of particular importance with respect to the low

\*. Corresponding author. E-mail: address: bahram330ghorbani@gmail.com (B. Ghorbani) temperature end of the cascade system, where a very substantial amount of power is wasted by this inherent inefficiency in such a cascade system [1].

Minimizing the work consumed by the refrigeration cycle is the most effective measure for reducing the cost of products of sub-ambient chemical processes. A recent advancement has been the introduction of mixed working fluids in refrigeration systems in place of pure working fluids. Refrigeration systems, known as Mixed Refrigerant Cascades, which can reach temperatures as low as 120 K, are under development in many parts of the world. This refrigeration method, with a multi-component mixture, has demonstrated high performance in low temperature ranges [2].

Nowadays, the MRC is widely used in commercial natural gas liquefaction fields whose liquefaction capacities are very large. The simplification of the compression and heat exchange processes in such a cycle may offer the potential for reduced capital expenditure, in comparison with conventional cascade cycles [3,4].

The concept of using a mixture as a refrigerant

has been around for a long time. The mixed refrigerant cycle was patented by Podbeilniak [5], which utilizes three stages of throttling, and a single compressor in a closed cycle. Thereafter, several MRCs were patented regarding gas liquefaction and separation applications [1,6-9]. Also, many investigations are available in open literature on the performance of MRCs in different low temperature applications [2,10-13].

The fundamental aspects of mixed refrigerant processes, though very innovative, have not received due attention in the open literature, in view of commercial interests. It is difficult to piece together the existing information to choose an appropriate process and an optimum composition for a given application. Venkatarathnam [14] has recently reviewed the fundamental aspects of mixed refrigerant processes, their advantages, the methods for optimizing the refrigerant composition and the performance of different processes with different nitrogen-hydrocarbon mixtures.

In sub-ambient processes, the design of refrigeration systems is a major concern for energy consumption and capital investment. The synthesis and optimization of MRCs for low temperature processes is complex, due to the large number of design options. In the design of mixed refrigeration systems for chemical processes, the main key issues are: composition of the mixed refrigerant, operating pressures (suction and discharge pressure of compressors), cycle configuration, and heat integration between refrigeration systems and process streams to achieve close matching of the hot and cold composite curves. There is no research in the literature on the designing of MRCs for low temperature industrial chemical processes using a systematic methodology that includes all the above mentioned aspects. The objective of this work is to present a methodology with graphical and numerical tools for the analysis, design and optimization of MRC for complex low temperature processes.

#### 2. Design of MRC with a given configuration

In the current paper, we concentrate on MRC design for complex low temperature chemical processes. As a typical example of low temperature processes, we have chosen an ethylene process (olefin plant). In an ethylene recovery process, a feed gas, comprising hydrogen, methane, ethane, ethylene, propane, propylene, and minor amounts of other light components, is compressed, cooled, and partially condensed in single stage condensers or, alternatively, in one or more dephlegmators, which imparts several stages of separation during the condensation step. The condensate is separated from lighter gases and is passed to one or more demethanizer columns, which recover light gas overheads comprised of chiefly methane and hydrogen, and a bottoms stream rich in  $C_2$  and  $C_3$ hydrocarbons. This hydrocarbon stream is further fractionated to yield a high purity ethylene product, an ethane-rich byproduct, and a stream of  $C_3$  and heavier hydrocarbons.

Essentially, all olefin plants use an ethylenepropylene cascade refrigeration system to provide the major portion of refrigeration required in the olefin plant. Most of the propylene (high level) refrigeration is utilized at several pressure/temperature levels in the initial feed precooling and fractionation sections of the plant to cool the feed from ambient temperature to about -35°C and to condense the ethylene refrigerant at about -30°C. Similarly, the ethylene (low level) refrigeration is utilized at several pressure/temperature levels in the cryogenic section of the plant to cool the feed from -35°C to about -100°C, in order to condense the bulk of the ethylene in the form of liquid feeds to a demethanizer column, and, in the demethanizer column, an overhead condenser at about -101°C to provide reflux to that column. Refrigeration below -101°C, to condense the remaining ethylene from the feed, is provided primarily by the work of expansion or the Joule-Thomson expansion of rejected light gases,  $H_2$ and methane, and/or by vaporization of the methane refrigerant which has been condensed by the ethylene refrigerant. The work of expansion or Joule-Thomson expanded gases is normally used as fuel and consists primarily of the overhead vapor from the demethanizer column (mostly methane) and any uncondensed feed gas (mostly  $H_2$  and methane) which is not processed in the  $H_2$  recovery section of the olefin plant.

The power consumption is always high in an olefin plant refrigeration system. Therefore, minimizing the work consumed by the refrigeration cycle is the most effective measure to reduce production costs in olefin plants.

# 2.1. Description of cascade refrigeration system

The cascade refrigeration system of the olefin plant analyzed in this study consists of ambient cooling water at near ambient temperature, closed cycle propylene and ethylene systems, and Joule-Thomson expansion of rejected light gases from the demethanizer column overhead vapor, and uncondensed feed gas in the  $H_2$ recovery section of the plant. Propylene refrigeration is utilized at several temperature levels (+5, -20)and  $-35^{\circ}C$ ) to cool and heat the feed in the initial fractionation sections of the plant. Similarly, the ethylene refrigeration is utilized at several temperature levels  $(-65^{\circ}C \text{ and } -101^{\circ}C)$  to cool the feed in the cryogenic section of the plant. Detailed description of the ethylene-propylene cascade refrigeration system has been presented in our previous work [15].

Figure 1 shows the flow diagram of an ethy-



Figure 1. Flow diagram of ethylene refrigeration cycle.

lene refrigeration cycle. Ethylene refrigerant vapors discharged from the compressor are desuperheated and condensed in E-504, E-505, E-506 and E-507, utilizing cooling water and 5°C, -20°C and -35°C liquid propylene refrigerant, respectively. As seen from Figure 1, in addition to the ethylene refrigerant, the cooling potential of the hydrogen rich gas, tail gas and regeneration gas streams has been used for cooling the feed stream in the heat exchanger, E-305. These cold process streams are provided by the Joule-Thomson expanded light overhead product from the demethanizer column and any hydrogen and methane which is not processed in the hydrogen recovery section.

Figure 2 shows the refrigeration system matched against the GCC (Grand Composite Curve) of the



Figure 2. GCC of the refrigeration system analyzed in this work [15].

olefin plant separation process analyzed in this Sometimes, the refrigeration levels are work [15]. fitted against a nearly flat portion of the GCC (e.g. propylene refrigeration levels in Figure 2). In this case, the pure refrigerants and azeotropic mixtures are the best options because of isothermal vaporization in the evaporators. Sometimes, the refrigerant level needs to be fitted against a slope of the GCC, as ethylene refrigeration levels shown in Figure 2. In this case, there is a degree of freedom in choosing the level of refrigeration [3]. For example, the GCC of the separation process analyzed in this work shows that having multiple stage evaporations for an ethylene refrigeration cycle makes the average temperature difference between the process streams and the refrigerant small. This results in smaller exergy destruction in the E-308 and E-305 evaporators, since the greater the temperature difference, the greater the exergy destruction. As the number of evaporation stages increases, the exergy destruction decreases. However, adding more stages means additional equipment cost, and more than two stages for an ethylene cycle in an olefin plant is not justified [16].

# 2.2. Developed MRCs for providing low level refrigeration

Going from the two-stage evaporation (including  $-65^{\circ}$ C and  $-101^{\circ}$ C levels) in the ethylene cycle, as shown in Figure 2, to a multiple-stage one, saves power, with an additional level of complexity, because of the



Figure 3. Mixed refrigerant cycle (configuration A).

restricted working range over which it can operate. The working range of refrigerant fluids can be extended and modified by using a mixture rather than a pure component. An MRC uses a mixture as a refrigerant instead of a pure refrigerant. Unlike pure refrigerants, the temperature and vapour and liquid composition of non-azeotropic mixtures do not remain constant at constant pressure as the refrigerants evaporate or condense [3].

Figures 3 and 4 show two flow-diagrams of MRCs (A and B configurations) developed in this section for providing low level refrigeration in place of the pure ethylene refrigeration cycle shown in Figure 1. Details of these cycles have been presented in our previous work [17]. Conditions and composition of feed, reflux and cold process streams in these flow-diagrams are similar to the pure ethylene refrigeration cycle.

# 2.3. Thermophysical properties of mixed refrigerant

In this paper, a mixture of hydrocarbons (propane, ethane, methane) and nitrogen is used to provide the desired refrigerant characteristics for the specific refrigeration demand in the MRCs. The difficulties in the design of MRC mainly come from two sources. First, the complex nature of the thermodynamic and physical properties of the mixtures makes the consumption of MRCs expensive and highly non-linear. Second, the small temperature approach between the hot and cold composite curves in multi-stream heat exchangers (the profiles of evaporation and condensation), and the wide temperature range. This not only increases the difficulty of modelling for the problem, but also adds to the non-linearity when carrying out optimization [16]. Therefore, an accurate prediction of the phase equilibrium for vapour-liquid ratios, and values of enthalpy and entropy, is essential for the mixtures.

### 2.3.1. Vapour-liquid equilibrium calculations

The equilibrium condition for every component of a two-phase mixture is expressed by the equality of fugacities. For a multicomponent refrigerant, the equilibrium criterion is given by:

$$f_i^V = f_i^L, \qquad i = 1, 2, \cdots, n,$$
 (1)

where:

- $f_i^V$  fugacity of component *i* in vapour phase mixture;
- $f_i^L$  fugacity of component *i* in liquid phase mixture ;
- *n* number of components in mixture.

The fugacity coefficient, defined as the ratio of fugacity to pressure, of each component in any phase is related to pressure, temperature and volume by the following generalized thermodynamic relationship [18,19]:

$$\ln \phi_i = \frac{1}{RT} \int_V^\infty \left[ \left( \frac{\partial P}{\partial n_i} \right)_{T,V,n_{j \neq i}} - \frac{RT}{V} \right] dV - \ln Z,$$



Figure 4. Mixed refrigerant cycle (configuration B).

$$i = 1, 2, \cdots, n, \tag{2}$$

where:

- V total volume of n moles of the mixture;
- $n_i$  number of moles of component i;
- Z compressibility factor of the mixture.

The fugacity coefficient can be calculated by an equation relating pressure, temperature, volume and compositions, that is, an equation of state. In general, any equation of state which provides reliable volumetric data over the full range of the above integral can be used to describe the fluid phase behaviour. The simplest and most highly successful equation is the semi-empirical two-parameter cubic equation, such as the Peng-Robinson and Soave-Redlich-Kwong equations [18]. In the present work, the Peng-Robinson equation of state has been used in calculation of the phase equilibrium. The Peng-Robinson equation of state is given as follows [19,20]:

$$P = \frac{RT}{v-b} - \frac{a\alpha}{v(v+b) + b(v-b)}.$$
(3)

Imposing classical critical point conditions on Eq. (3) and solving for parameters a and b yields:

$$a = \Omega_a \frac{R^2 T_C^2}{P_C}, \qquad \Omega_a = 0.45724,$$
  
$$b = \Omega_b \frac{RT_C}{P_C}, \qquad \Omega_a = 0.07780, \qquad (4)$$

where the subscript, C, refers to the values at the critical point, and R is the gas constant.

The temperature dependent parameter,  $\alpha$ , is defined by:

$$\alpha = \left[1 + (0.3796 + 1.5422\omega - 0.2699\omega^2) \right]^2,$$
(5)

where  $\omega$  is the acentric factor.

To apply equations of state to multicomponent systems, their parameters for the mixtures have to be defined by employing some mixing rules. The random mixing rule has been recommended for all two-constant cubic equations of state. It defines the constant and temperature dependent parameters of the Peng-Robinson equation of state for mixtures as:

$$P = \frac{RT}{v - b_m} - \frac{(a\alpha)_m}{v(v + b_m) + b_m (v - b_m)},$$
  

$$(a\alpha)_m = \sum_i \sum_j \left[ y_i y_j \sqrt{a_i a_j \alpha_i \alpha_j} (1 - k_{ij}) \right],$$
  

$$b_m = \sum_i [y_i b_i],$$
(6)

where the indices, i and j, denote the components, and  $k_{ij}$  is the binary interaction coefficient. Eq. (6) can be rewritten as:

$$Z_m^3 + (B_m - 1)Z_m^2 + (A_m - 3B_m^2 - 2B_m)Z_m - (A_m B_m - B_m^2 - B_m^3) = 0,$$
$$A_m = \frac{(a\alpha)_m P}{(RT)^2},$$
$$B_m = \frac{b_m P}{RT},$$
(7)

where Z is a constringent factor, A and B are the coefficients relating to the gas state parameters and m denotes the mixture.

### 2.3.2. Thermodynamic properties calculation

The Lee-Kesler equation of state is an accurate general method for prediction of the thermodynamic properties of non-polar mixtures [20]. In this work, the mixed refrigerant consists of methane, ethane, propane and nitrogen. Therefore, the Lee-Kesler equation of state has been used in calculation of the enthalpy and entropy of the mixed refrigerant. This model is expressed as:

$$Z = Z^{(0)} + \frac{\omega}{\omega^{(r)}} \left( Z^{(r)} - Z^{(0)} \right), \tag{8}$$

where  $\omega$  is an acentric factor, and 0 and r denote the relevant parameters of simple and reference liquids.

### 2.4. Optimization algorithm

Many factors influence the performance of a certain MRC, for instance, operating pressures of the cycle (suction and discharge pressures of cycle compressor), the temperature of the refrigerant before expansion, and the mole fraction of mixed refrigerant components such as nitrogen, methane, ethane, and propane, etc. In this work, the optimization problem consists of the determination of the optimum parameter values that minimize the power consumption. The objective function is:

$$\min f(x_1, x_2, \cdots, x_n) = W, \tag{9}$$

where W is the power consumption of the mixed refrigerant compressor and  $x_i$  denotes operating parameters, such as the composition of the refrigerant, and the suction and discharger pressures of the compressor, etc. In fact, it is necessary to take into account all factors, such as initial cost, power consumption, plant area, and the simplicity of the process, etc., but many of these factors are not purely technical [12]. In this paper, only the power consumption is considered as the optimization objective. The constraints are as follows:

• The sum of the mole fractions of the mixed refrigerant is 1.

- The temperature of the mixed refrigerant at the compressor inlet is higher than its dew point.
- The temperature difference between the hot and cold streams cannot be negative.

In this section, we have proposed a systematic design method to find the optimum values of operating pressure and refrigerant composition, which minimize the power consumption of MRC with a given configuration. The basic idea is to find a set of refrigerant compositions that give minimum power consumption under given pressure levels (high and low operating pressures of MRCs) and satisfy all above mentioned constraints. Then, the pressure levels of MRC are changed in limited ranges defined by the user, and the procedure of finding the best refrigerant composition is repeated iteratively. Figure 5 explains the methodology.

As seen from Figure 5, to take the best solution and avoid being trapped in local optima, a two-phase hybrid method has been developed. The first phase is explorative, employing an Enumerative method to identify promising areas of the search space. The best solution found by the enumerative method is then



Figure 5. Proposed systematic design method for optimal selection of refrigerant composition and operating pressures of MRC with a given configuration.

refined using a pattern search method during a subsequent exploitative phase. We selected hybridization of global and local search algorithms to produce high quality optimal solution, although computational time is relatively expensive.

It should be mentioned that an important feature of the proposed method is to ensure heat integration between the refrigeration system and process streams. It is guaranteed by combining the cold process streams (hydrogen rich gas, tail gas and regeneration gas streams in this study) and cold refrigerant streams as a cold composite curve, and also combining the hot process streams (feed and reflux streams in this study) and warm refrigerant as a hot composite curve.

The domain of the optimized mixture composition is restricted by consideration of a suitable mixed refrigerant for the cracking of liquid or gaseous charges in the olefin plant. Good results have been achieved with a refrigerant having the following composition, based on mole fraction [7]:

- $N_2: < 3\%;$
- CH<sub>4</sub>: 5% to 30%;
- $C_2H_6$ : 30% to 60%;
- $C_3H_8$ : 10% to 60%.

The temperature at the outlet of the subcooler shown in Figures 3 and 4 is assumed constant and to be  $-90^{\circ}$ C.

#### 2.5. Simulation results and discussions

The mixed refrigerant cycles have been simulated at steady-state condition. Simulation results have been obtained based on 75% isentropic efficiency for the compressors.

Table 1 obtains the solution found by the enumerative method to take the best solution in the neighborhood of the global optimum.

Table 2 obtains the refined solution using the direct search method, which minimizes the power consumption of MRCs shown in Figures 3 and 4. The simulation results reveal that each MRC configuration has its own optimal mixture composition and optimal high and low operating pressures.

As seen from Figures 1, 3 and 4, it is obvious that the condensers of pure ethylene and MRCs are affected by the propylene evaporators, including E-505, E-506 and E-507, which should extract towards the outside a major portion of the heat extracted by the ethylene cycle and MRCs. This leads to the need to provide a large refrigerating circuit working with propylene, which requires large compressor power consumption. Table 3 presents the key parameters of a pure-ethylene cycle and two optimized MRCs based on the systematic design method explained in the previous section.

It can be found from this table that configuration B behaves thermodynamically in a more favorable manner than the pure-ethylene cycle, thereby, making it possible to achieve substantial power saving for providing the same refrigeration duty. The shaft work

 Table 1. The best solution in the neighborhood of global optimum for MRCs shown in Figures 3 and 4 using enumerative method.

Configuration	$P_{\min}$	$P_{ m max}$	Propane	$\mathbf{Ethane}$	Methane	$\mathbf{Nitrogen}$
Configuration	$(\mathbf{kPa})$	$(\mathbf{kPa})$	(mol%)	(mol%) $(mol%)$		(mol%)
А	180	1450	32	35	32	1
В	200	1250	36	34	29	1

Table 2. Optimal operating conditions for MRCs shown in Figures 3 and 4 using pattern search method.

Configuration	$P_{\min}$	$P_{\max}$	Propane	Ethane	Methane	Nitrogen
	(kPa)	(kPa)	(mol%)	(mol%)	(mol%)	(mol%)
А	180	1450	32.30	35.19	32.50	0.01
В	200	1250	36.72	33.85	28.77	0.66

Table 3. $($	Comparison	between th	ie key	parameters of	of pure e	thylene	and optimiz	ed MRCs.
--------------	------------	------------	--------	---------------	-----------	---------	-------------	----------

Type of cycle	$P_{ m max}$ (kPa)	$P_{ m min} \ ( m kPa)$	Power consumption of cycle compressor (kW)	Power consumption of Propylene cycle compressor (kW)	Total power Consumption (kW)
Pure ethylene	2020	108	1664	5299	6963
Configuration A	1450	180	1843	4089	5932
Configuration B	1250	200	1489	4118	5607

of configuration B is calculated to be 1489 kW, which is 175 kW lower than that required by a pure ethylene refrigeration cycle.

The results show that MRCs can improve the thermodynamic performance of refrigeration systems in the case of using optimal mixture compositions, optimal high and low operating pressures and also proper arrangement of the cycle components (cycle configuration).

#### 3. Optimal arrangement of MRC

A refrigeration cycle is a chain of various equipment including compressors, condensers, evaporators and expansion devices, each of which takes on a particular thermodynamic process duty in the cycle, which eventually results in production of the required refrigeration.

From the data analysis of Tables 1 and 2, it is evident that configuration of MRC is an effective parameter in power consumption. In spite of the simplicity of the MRCs machinery configuration in comparison with a conventional cascade cycle, it is possible to imagine different configurations, providing the process required refrigeration, for these cycles. But the question here is how to find a configuration taking advantage of the existing complicated interactions between low temperature processes and refrigeration cycles, so that the maximum efficiency and the best functionality are guaranteed.

In order to answer the above question, a background section is included prior to introducing a methodology for finding the optimal MRC configuration for providing refrigeration in a certain low temperature process.

#### 3.1. Background

In the previous section, two MRCs were developed for a typical olefin plant utilizing a mixture of methane, ethane, propane and nitrogen, as the cycle working fluid to replace the pure ethylene refrigeration cycle. Also, a systematic design method to meet the objective of minimum shaft work in the compressor was suggested to optimize the major parameters of each MRC, including high and low operating pressures and refrigerant mixture composition.

The strength of the suggested method is that it combines mathematical programming and thermodynamic methods, which can generate optimal design solutions and a good understanding of design problems, while mathematical programming also produces an optimal solution for the cycle. Thermodynamic programming draws the composite curves of the MRC processes and, therefore, expresses the evaluation of the solution procedures in a visual way, so that the designer has understanding and confidence in the solution.



Figure 6. The balanced composite curve of configuration A mixed refrigerant cycle.



**Figure 7.** The balanced composite curve of configuration B mixed refrigerant cycle.

For instance, Figures 6 and 7 illustrate the balanced composite curves for two MRCs after optimizing the key parameters using an enumerative method. It should be noticed that in these curves, the thermal integration of MRC with low temperature process streams is accomplished by combining the process hot and cold streams and those of the refrigerant.

As shown in Figures 3 and 4, the heat of condensing from the compressed Mixed Refrigerant (MR) is transfered to cooling water in condenser E-504 and in the pure-propylene refrigeration cycle. To observe the impact of MRC on the precooling refrigeration cycle (propylene cycle in this study), the balanced composite curves of the MRC can be modified, so that they become indicative of the rejected heat from the compressed MR. Figures 8 and 9 indicate the modified composite curves for two optimized MRCs, called a Grand Composite Curve (GCC). Dashed lines in these figures represent condensation of the MR in the cooling water and liquid propylene refrigerant condensers. It can be derived from these figures that the hot and cold



**Figure 8.** The GCC of configuration A mixed refrigerant cycle.



**Figure 9.** The GCC of configuration B mixed refrigerant cycle.

composite curves in configuration A are matched better than those in configuration B.

The main reason for differences in the GCC of MRCs with various configurations lies in the diverse possible configuration and arrangement of these cycles and their related equipment. In other words, it can be stated that having optimized the design variables of MRCs, their GCCs are suitable criteria in comparing various configurations of these cycles. These curves can be regarded as a qualitative criterion to measure cycle deviation from the desired condition (close matching between hot and cold composite curves).

The composite curves can be redrawn by replacing the temperature with the Carnot factor ( $\eta = 1 - [T_0/T]$ ), resulting in the ECC (exergy composite curves) as shown in Figures 10 and 11. The area between the curves in these diagrams represents the exergy loss in the utility exchangers [21]. The question, here, is whether the changes in the area enclosed in ECC, given for MRCs, due to change in the cycle arrangement, equal the changes in cycle power consumption.



**Figure 10.** Exergy composite curves of configuration A mixed refrigerant cycle.



Figure 11. Exergy composite curves of configuration B mixed refrigerant cycle.

In response to the above question, consider Tables 4 and 5 and compare the difference between the values of the enclosed area between MR condensation and liquid propylene refrigerant curves (light grey colored region), and also the hot and cold composite curves (dark grey colored region) in Figures 10 and 11 with the difference between the power consumption of MRCs and the propylene cycle (precooling cycle) for configurations A and B, optimized using the enumerative method.

From the data analysis of Tables 4 and 5, it is evident that values of the difference between power consumption and the one between the area of enclosed regions for configurations A and B are close to each other. This difference can be minimized computing the exergy losses in throttling values and compressors, along with adding them to exergy loss in heat exchangers (the area of the enclosed region). Therefore, ECC can be used as a tool to establish proportionality between the changes in the configuration of MRC and the change in power consumption of the cycle.

In low temperature processes with a pure refrigerant cycle, ECC can be used as a shaft work

Row	$\mathbf{MRC}$	Power consumption of	Area of light grey	
No.	configuration	Propylene cycle compressor (kW)	colored region $(kW)$	
1	Configuration A	4089	1061	
2	Configuration B	4118	1093	
Differen	ce between the values	20	3.0	
of ro	ws No. 1 & 2 (kW)	29	52	

**Table 4.** Comparison of the difference between the area of light grey colored regions in Figures 10 and 11 with the difference between power consumption of propylene cycles.

**Table 5.** Comparison of the difference between the area of dark grey colored regions in Figures 10 and 11 with the difference between power consumption of MRCs.

Row	MRC	Power consumption of	Area of dark grey	
No.	configuration	$\mathbf{MRC}\ \mathbf{compressor}\ (\mathbf{kW})$	colored region $(kW)$	
1	Configuration A	1876	5044	
2	Configuration B	1528	4841	
Differen of ro	uce between the values ws No. 1 & 2 (kW)	203	348	

targeting tool to directly estimate the change in power consumption of the cycle (owed to modifications and improvements in the cycle configuration) without going through detailed refrigeration calculations [21]. In other words, ECC can assist the designer in finding the best refrigeration system configuration.

MRC uses a mixture as a refrigerant instead of a pure refrigerant. Unlike a pure refrigerant cycle, graphical targeting approaches, such as ECC and GCC, cannot be used directly to optimize the MRC configuration, because the optimization requires adjustments to the refrigerant composition. But, MRC features a simpler machinery configuration in comparison with a pure refrigerant multiple stage cycle. Therefore, ECC and GCC charts can be used as a graphical tool for feeling and understanding MRC behavior. These curves tend to provide "suggestive elements" for improving the cycle configuration.

Let us consider the ECC and GCC charts of the MRCs shown in Figures 3 and 4. As can be seen from Figures 8 and 9, the gap between the hot and cold composite curves for configuration A is larger in the temperature range of  $-60^{\circ}$ C to  $-90^{\circ}$ C in comparison with configuration B. It is caused by passing MR immediately after the expansion valve into the subcooler of configuration A, which imposes a great temperature difference, along with thermal exchange, consequently leading to a rise in exergy loss (Figure 10). Also, configuration B cycle has been equipped to multistream heat exchangers (No. 1 and No. 2 heat exchangers shown in Figure 4), which results in a better matching between the hot and cold composite curves (Figure 11).

# 3.2. Proposed method for optimizing MRC configuration

Due to the power and capabilities of GCC and ECC charts in indicating room for further improvement in the MRC process, the systematic design method proposed for optimal selection of refrigerant composition and operating pressures (Figure 5) has been extended to cover the cycle's configuration optimization.

Figure 12 depicts the methodology. It comprises two main parts: Basic design phase and detailed design phase. The procedure commences from an initial guess for MRC configuration.

In the basic design phase, by employing the enumerative method, the best refrigerant composition and operating pressure of the cycle are found under conditions imposed by a low temperature process. Drawing the GCC and ECC charts is the next step. By considering the match between hot and cold composite curves of the initial guess, room for further improvements is indicated. Then, the designer can modify the cycle configuration based on an understanding of MRC behavior, heuristics and judgment. The procedure stops when no further improvement is possible.

In the detailed design phase, the MRC found from the basic phase is fed to a direct search method to explore further possibilities for reducing power consumption.

To cut down the computational time of the explorative stage in the basic design phase, the search space firstly is discriminated by using coarse steps. Then, in the next iterations, by providing a feel of the cycle behavior as well as restricting the domain of the search space, the discrete steps can be fined.

An illustrative example of how to utilize the pro-



Figure 12. Proposed design method to give optimal arrangement of MRC.

posed method to achieve optimal MRC configuration has been presented in the next section.

#### 3.3. Case study

In previous sections, two MRCs have been designed on the basis of the characteristics of the olefin plant cryogenic section in place of pure ethylene refrigeration. By considering the ECC and GCC charts of these cycles, it was concluded that using multistream heat exchangers in the cycle configuration will lead to a better matching between hot and cold composite curves, resulting in lower cycle power consumption. Indeed, by using suitable multistream heat exchangers, the distribution pattern of hot and cold streams will be then out of the designer's scope of work, and the burden will be on the optimization algorithm of the systematic design method. Knowing this idea significantly aids the cycle configuration optimization. As an example, in MRC with configuration B, which is shown in Figure 4, the designer has proposed a certain arrangement for heat exchangers that imposes a distribution plan of hot and cold streams. Consequently, the systematic design method would only be able to optimize the refrigerant composition and operating pressures. To overcome this problem, the configuration depicted in Figure 13 has been suggested, which obtains a better match between hot and cold streams.

Table 6 compares the optimized variables found by the enumerative method for configurations B and C.

The results show that by altering the cycle configuration to take better advantage of the heat integration between MRC and process streams in a multistream heat exchanger, a drastic reduction in cycle power consumption (239 kW) has occurred.

Figures 14 and 15 illustrate the GCC and ECC charts of the configuration C mixed refrigerant cycle, respectively. By comparing the GCC in configurations B and C (Figures 14 and 9), a considerable improvement between the hot and cold composite curves can be observed in configuration C.



Figure 13. A mixed refrigerant cycle suggested to obtain a better match between hot and cold streams in multistream heat exchanger (configuration C).

Table 6. The optimized variables for MRCs shown in Figures 4 and 13 using enumerative method.

Configuration	$P_{\min}$	$P_{ m max}$	Propane	Ethane	Methane	Nitrogen	Power consumption
Configuration	$(\mathbf{kPa})$	$(\mathbf{kPa})$	(mol%)	(mol%)	(mol%)	(mol%)	$(\mathbf{kW})$
В	200	1250	36	34	29	1	1528
$\mathbf{C}$	260	800	30	45	24	1	1239



Figure 14. The GCC of configuration C mixed refrigerant cycle.

Figures 14 and 15 indicate that although the proposed design method has caused the hot and cold streams in the heat exchanger to close each other, there is still considerable temperature difference between hot and cold streams around the pinch point in the heat exchanger network. In order to cut down the temperature difference along the heat exchanger, it is possible to distribute the thermal exchange existing between cold and hot streams among several cascade heat exchangers.

Figure 16 illustrates a two-stage MRC proposed in this paper instead of a single-stage cycle (configuration



Figure 15. The ECC of configuration C mixed refrigerant cycle.

C) in order to decrease the power consumption. In this cycle, the amount of refrigerant flow in each heat exchanger is different, adding one more degree of freedom in the design of the MR systems. This extra degree of freedom creates opportunities to achieve a more efficient design, but it also causes more complexities in the modeling of MR systems. The process conditions are set the same as in the two MRCs developed in the previous section.

It is reasonable to assume that all outlet streams from the cold end multistream have equal temperatures [16]. This temperature, which plays a significant



Figure 16. A two-stage MRC (configuration D).



Figure 17. The ECC of two-stage MRC, intermediate temperature: -65 °C.

role in the refrigeration cycle, is called the cycle intermediate temperature. Intermediate temperature can alter the shape of the hot and cold composite curves. Figure 17 shows the ECC charts for the MRC shown in Figure 16.

Intermediate temperature can alter the shape of the hot and cold composite curves. In this study, the change to intermediate temperature has been done manually, following observations and heuristics. Table 7 compares the results derived from optimization of the parameters of the two-stage refrigeration cycle with other cycles.

It can be found from this table that despite the more complex two-stage refrigeration cycle, in comparison with configuration C, there is no drastic reduction in cycle power consumption. Thus, it can be deduced that the configuration C cycle is the best substitute for the refrigeration cycle of the pure ethylene refrigerant in the olefin plant analyzed in this study.

It should be noted that lower power consumption can be expected if the number of stages is increased, but inevitably results in greater complexity and difficulty of control. Moreover, the effect of reducing power consumption by increasing the number of stages is progressively diminished.

#### 4. Conclusion

In this paper, a systematic design method for optimal selection of refrigerant composition and operating pressures of MRC with a given configuration has been presented. Based on the success of the proposed systematic method for designing MRC with a given configuration, the method is extended to give the optimal arrangement of cycle components. The essence of the extended method is the proper combination of pinch and exergy analysis in a visual way. Thus, the causes of inefficiency in the mixed refrigerant cycle configuration can be quickly identified. Based on these insights, the designer can confidently evolve better designs and introduce ideas for improving the cycle arrangement.

The solution that gives the lowest shaft work requirement may incur an extra large heat transfer area and thus capital costs. Giving better guidelines for picking the most economic solution and minimizing the total cost can be achieved by combining systematic design method proposed in this paper with a design

Table 7. The key parametrs of various MRCs optimized using enumerative method.

Configuration	$P_{ m min}$ (kPa)	$P_{ m max}$ (kPa)	Propane (mol%)	Ethane (mol%)	$egin{array}{c} { m Methane} \ ({ m mol}\%) \end{array}$	$egin{array}{l} { m Nitrogen} \ ({ m mol}\%) \end{array}$	Power consumption (kW)
В	200	1250	36	34	29	1	1528
С	260	800	30	45	24	1	1239
D	140	600	43	41	15	1	1230

method for synthesis of a multistream heat exchanger, which is the topic for future works.

### References

- 1. Gaumer, L.S. and Newton, C.L. "Combined cascade and multicomponent refrigeration system and method", US patent 3763658 (1979).
- Gong, M., Wu, J. and Luo, E. "Performance of the mixed-gases Joule-Thomson refrigeration cycles for cooling fixed-temperature heat loads", *Journal of Cryogenics*, 44, pp. 847-857 (2004)
- 3. Smith, R., Chemical Process Design and Integration, John Wiley & Sons, New York (2004).
- Flynn Thomas, M., Cryogenic Engineering, Marcel Dekker, New York (2005).
- Podbeilniak, W.J. "Art of refrigeration", US patent 2041725 (1936).
- Kleemenko, A.P. "One flow cascade cycle", Proceeding of the 10th International Congress of Refrigeration, 1, pp. 34-39 (1959).
- Becdeliever, C., Kaiser, V. and Paradowski, H. "Method of arrangement for processing through low temperature heat exchanges in particular for treating natural gases and cracked gases", US patent 4072485 (1978).
- 8. Howard, L. and Rowles, H. "Mixed refrigerant cycle for ethylene recovery", US patent 5379597 (1995).
- Wei, VT. "Ethylene plant refrigeration system", US patent 2002/0174679 A1 (2002).
- Gong, M., Wu, J., Luo, E., Qi, Y. and Zhou, Y. "Study of the single-stage mixed-gases refrigeration cycle for cooling temperature-distributed heat loads", *International Journal of Thermal Science*, 43, pp. 31-41 (2004).
- Alexeev, A., Thiel, A., Harberstoh, Ch. and Quack, H. "Study of behavior in the heat exchanger of a mixed gas Joule-Thomson cooler", *Advances in Cryogenics Engineering*, 45, pp. 307-315 (2000).
- Cao, W., Lu, X., Lin, W. and Gu, A. "Parameter comparison of two small-scale natural gas liquefaction processes in skid-mounted packages", *Journal of Applied Thermal Engineering*, 26, pp. 898-904 (2006).
- Remeljej, C.W. and Hoadly, A.F.A. "An exergy analysis of small-scale liquefied natural gas (LNG) liquefaction processes", *Journal of Energy*, **31**, pp. 1669-1683 (2006).
- 14. Venkatarathnam, G. "Cryogenic mixed refrigerant processes", Springer: International Cryogenic Monograph Series, New York (2008).
- Mafi, M., Mousavi Naeynian, S.M. and Amidpour, M. "Exergy analysis of multistage cascade low temperature refrigeration systems used in olefin plants", *International Journal of Refrigeration*, **32**, pp. 279-294 (2009).

- Lee, G.C. "Optimal design and analysis of refrigeration systems for low-temperature processes", PhD Thesis, UMIST, UK (2001).
- Mafi, M., Amidpour, M. and Mousavi Naeynian, S.M. "Development in mixed refigerant cycles used in olefin plants", Elsevier B.V.: Advances in Gas Processing, Volume 1: Proceedings of the 1st Annual Gas Processing Symposium, 1, pp. 154-161 (2009).
- Danesh, A. "PVT and phase behaviour of petroleum reservoir fluids", Elsevier B.V.: Development in Petroleum Science, 47, Third impression (2003).
- Ahmed Tarek, H., Equation of State and PVT Analysis: Application for Improved Reservoir Modeling, Gulf Publishing Company: Houston, Texas (2007).
- 20. Reid, R.C., Prausnitz, J.M. and Poling, B.E., *The Properties of Gases and Liquids*, McGraw-Hill, Fourth Edition (1987).
- Linnhoff, B. and Dhole, V.R. "Shaftwork targets for low temperature process design", *Chemical Engineer*ing Science, 4(8), pp. 2081-2091(1992).

#### **Biographies**

Mostafa Mafi received his BS degree in Mechanical Engineering from K.N. Toosi University of Technology, Iran, in 2002, his MS degree in Mechanical Engineering from Tabriz University, Iran, in 2004, and his PhD degree in Mechanical Engineering from K.N. Toosi University of Technology, Iran, in 2009. He is currently Assistant Professor of Mechanical Engineering at Imam Khomeini International University, Iran. His research interests include cryogenics systems, low temperature refrigeration systems, LNG plants, liquefaction and separation systems in petrochemical industries, airconditioning systems and energy saving. He has published more than 25 papers in refereed journals and conferences, and is a member of the Iranian Society of Heating, Refrigeration and Air Conditioning Engineers (IRSHRAE).

Bahram Ghorbani obtained his BS degree in Mechanical Engineering from the University of Mazandaran, Iran, in 2010, and his MS degree in Energy System Engineering from K.N. Toosi University of Technology, Iran, in 2012. His research interests are currently focused on energy saving in chemical processes, turbomachinery and propulsion systems.

Majid Amidpour received a BS degree in Chemical Engineering from Tehran University, Iran, and MS and PhD degrees in Process Integration and Energy Conservation from UMIST. His specific research interest is energy systems, and he has published more than 300 papers in this field in refereed journals and conferences.

He is on the managerial board of the Iran hydrogen and fuel cell association, member of the national energy committee and the national chemical engineering society, IChemE (1995-1998), AICHE (1996-1998). He is member of the editorial board of the Iranian Chemistry & Chemical Engineering Journal, and has supervised 10 PhD and more than 50 MS theses in fields related to energy systems. He is currently head of the Chemical Engineering Department in Azad University, Iran, and director of the MS joint degree program with Kingston University and Manchester University in the UK.

His professional experiences include head of energy and process optimization department in IR. R&D, research deputy in Iranian Research and Developing Center for chemical industry, head of IR.R&D for chemical industry, research consultant in Ministry of Industry (innovation and energy saving), research and also education deputy in Mechanical Engineering Faculty at K.N. Toosi University of Technology, head of energy system engineering, consultant Oil and Energy Ministries company's in Iran, power plant optimization in Iran power stations, referee of three international journal.

Seyed Mojtaba Mousavi Naeynian received his PhD degree in Mechanical Engineering from Moscow University, Russia, and is currently Associate Professor of Mechanical Engineering at K.N. Toosi University of Technology, in Iran. He has authored four books and numerous professional journal articles and supervised 5 PhD and more than 50 MS theses in fields related to refrigeration systems. His research interests include: Cryogenics and Refrigeration Systems, and he is an expert on chill space design for different applications. He is a member of Iranian Society of Heating, Refrigeration and Air Conditioning Engineers (IRSHRAE).