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A Novel Approach of -80°C Cascade Refrigeration System Using Non-Flammable Quaternary Refrigerants for Semiconductor Process Applications

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Abstract: Ultra-low temperature chillers have seen increasing demand with the advancement of semiconductor technology. Mixed refrigerant (MR) cascade refrigeration systems (CRSs) are widely utilized for their stability and high cooling performance at low temperatures. Extensive research has been conducted on optimizing MR, which has a significant impact on CRS performance. However, most previous studies have either fixed the system pressure or used the refrigeration effect as the sole performance indicator. This did not account for the potential of achieving higher performance with an optimal MR composition at the same target temperature. In this study, a detailed parametric analysis was performed to investigate how the mass fractions of high-, mid-, and low boiling point refrigerants affect the coefficient of performance (COP) and exergy in ultra-low temperature CRSs without fixing the suction pressure. The analysis revealed that at the point of maximum COP, the refrigeration effect was relatively low, highlighting the limitations of using the refrigeration effect alone as a performance indicator. Additionally, COP was found to inversely correlate with total exergy destruction. As cascade temperature increases, COP tends to decrease, emphasizing the need for appropriate cascade temperature selection for MR CRS performance. This study introduces a novel approach to optimizing MR composition under various operating conditions, contributing to the advancement of ultra-low temperature CRSs.



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Keywords: ultra-low temperature; mixed refrigerants; coefficient of performance; exergy; semiconductor technology

1. Introduction

Owing to the surging demand for semiconductors with the advent of the AI era, the semiconductor industry has been expanding worldwide in recent years [1]. This growth has intensified the need for advanced manufacturing technologies, particularly those that enhance precision and performance. A crucial component in this evolution is the etching process, where precise temperature control is essential for accurate pattern formation and overall device quality. Recent advancements have introduced ultra-low temperature etching processes, especially for high-performance devices such as NAND flash memory. These processes operate at temperatures ranging from -70 to -80°C , significantly improving etching precision and speed by enhancing the uniformity of the etching reaction and enabling the formation of intricate patterns. As a result, there is a growing demand for ultra-low temperature chillers that can achieve and maintain these extreme conditions. These chillers are critical not only for removing the heat generated during the development of highly integrated semiconductor technologies but also for ensuring the high performance and reliability of advanced semiconductor devices [2,3]. Given the ongoing advancements in semiconductor technology, the demand for high-performance ultra-low temperature chillers is expected to continue rising, highlighting their essential role in the future of semiconductor manufacturing.

Typical technologies for the ultra-low temperature refrigeration of chillers include two-stage compression and the cascade refrigeration system (CRS). The CRS has been commonly used as it can reach lower temperatures in a stable manner compared with two-stage compression [4]. In the ultra-low temperature range of CRSs, which is from -50 to -80 °C, R-404A has been mainly used in the high-temperature cycle (HTC), and the refrigerants R-23 and R508B have been used in the low-temperature cycle (LTC) [5,6]. L.H.P. Massuchetto et al. [7] said that, so far, only the use of pure refrigerants has been studied even though there are various CRS optimization studies on exergy and economic efficiency analyses, which indicate that the use of mixed refrigerants (MR) is a promising option for obtaining a higher coefficient of performance (COP).

Subsequently, a number of studies revealed that the application of MR to CRSs can reduce the compressor power consumption and increase the COP by decreasing the compression ratio at ultra-low temperatures [8]. J. Lee et al. [9] reported that high power consumption is required to maintain an ultra-low temperature when the surrounding environment is not negligible based on the second law of thermodynamics and that the use of MR can significantly improve the low thermodynamic efficiency of ultra-low temperature chillers because it significantly decreases the operating pressure and increases the refrigeration effect compared with the use of pure refrigerants at ultra-low temperatures. They applied an MR composed of nitrogen, methane, ethane, and propane in the precooling cycle and an MR composed of nitrogen and neon in the main cycle and then compared and analyzed the COP and exergy efficiency of the system according to the number of heat exchangers. G. Di Nicola et al. [10] used R744 and HFC blends as low-temperature fluids and conducted a thermodynamic analysis of a CRS with various MR. Nasruddin et al. [11] analyzed the energy and economic efficiency for the optimization of CRSs using C₃H₈ in the HTC and a C₂H₆/CO₂ mixture in the LTC. A. Rozhentsev et al. [12] reported that the use of MR can lead to a high performance with a proper compression ratio in the low-temperature region of JT cooling systems due to the unique thermodynamic properties of MR in the 2-phase envelope. M. Gong et al. [13] compared and analyzed the system performance through an experiment in which a single refrigerant (R508B) and an MR composed of R170 and R116 were used in the LTC of ultra-low temperature CRSs. They revealed that the COP of the CRS that used MR was 10% higher than when a single refrigerant was used.

Many previous studies on performance improvement through the application of MR to CRSs were focused on the optimization of MR, as it has a significant impact on the system performance. T. Bai et al. [14] experimentally revealed that the composition ratio of MR significantly affects the system performance. As such, studies on MR composition ratio optimization have been actively conducted in various research fields related to low temperature, and some previous studies are as follows. N. Tzabar [15] expressed the isothermal refrigeration effect of a specific composition ratio as a performance indicator when the mole fraction of nitrogen/ethane and nitrogen/propane MR was changed under ultra-low temperature conditions under a high pressure of 2 MPa and a target temperature of 100 K. N.L. Narasimhan et al. [16] expressed the isothermal JT refrigeration effect of a specific composition ratio as a performance indicator at a high pressure of 17 bar and a low pressure of 3 bar using different N₂-hydrocarbon mixtures to identify the effect of the composition ratio of the MR on the performance of the JT cooler. P. Dorosz et al. [17] used the gross capacity, which has the same meaning as the isothermal JT refrigeration effect, of a specific composition ratio of MR as a performance indicator under the conditions of a high pressure of 20 bar and a low pressure of 1.1 bar. J. Lee et al. [9] selected the MR with the highest COP using the HYSYS SQP algorithm under the conditions of a CRS suction pressure of 1 bar and a maximum discharge pressure of 40 bar. Z. Zhao et al. [18] adjusted the operating pressure and the composition ratio of the MR using the genetic algorithm of the MATLAB optimization toolbox to maximize COP at a specific cold space temperature under the conditions of a CRS suction vapor quality of 1 and a maximum discharge temperature of 120 °C. Most of these studies, however, fixed the high and low pressures of the system at specific pressures and used the isothermal JT refrigeration effect

as a performance indicator for finding the optimal composition ratio of the MR. As these were studies conducted under limited pressure simulation conditions, they did not consider the possibility of obtaining the optimal MR composition ratio with a better refrigeration effect and higher COP at the same target temperatures by using other parameters, such as the temperature and quality, as simulation conditions instead of pressure.

In addition, C. Lee et al. [19] pointed out that flammable refrigerants were commonly used in MR JT compared with non-flammable refrigerants due to their larger isothermal enthalpy difference, but that there is an insufficient number of studies on non-flammable MR JT coolers despite the explosion or flammability risk of flammable refrigerants. T. Bai et al. [14] performed an experiment applying an MR composed of R170 and R290 with a low GWP in a JT cooler for refrigeration at $-60\text{ }^{\circ}\text{C}$, but the compression ratio was very high (21.8 to 26.8) under certain operating conditions. In addition, the refrigerants used were highly flammable as their ASHRAE safety group was A3. Y. Qin et al. [20] mixed R1234yf with R170, R41, and R1132a in the temperature range of -40 to $-60\text{ }^{\circ}\text{C}$ and compared the Linde-Hampson system COP and exergy efficiency of various MRs. They revealed that the COP and exergy efficiency were the highest when R1234yf was mixed with R1132a. Z. Zhao et al. [18] mentioned that it is still difficult to apply HFO-based refrigerants, such as R1234yf and R1234ze(E), to actual medium to large refrigeration systems due to their low cooling capacity at low temperatures caused by their high boiling points as well as high flammability. In addition, Y. Song et al. [21] pointed out that studies on ultra-low temperature CRSs have so far been developed mainly in the -50 to $-60\text{ }^{\circ}\text{C}$ target temperature range and that more studies on CRSs below $-80\text{ }^{\circ}\text{C}$ are required. There are strict flammability regulations for semiconductor processing chillers that require a target temperature of $-80\text{ }^{\circ}\text{C}$ or lower. As alternative refrigerants have their own limitations, such as efficiency degradation, high pressure, and flammability, comprehensive and practical research is required to improve the performance and efficiency by applying MR to CRSs at temperatures below $-80\text{ }^{\circ}\text{C}$.

Against this backdrop, the aim of this study is to analyze the COP and exergy of CRSs according to the mass fraction of a non-flammable MR at a cooling capacity of the semiconductor processing chiller of 8 kW at a target temperature of $-80\text{ }^{\circ}\text{C}$. Simulations were performed without fixing the suction pressure to provide a new optimal composition ratio strategy for selecting the MR composition ratio under various operating conditions of the system. There were many studies that mixed the composition ratio of MR by 0.1 or 0.5 units [19,22], but the simulation in this study was performed by mixing a four-component MR by 0.01 units for a more detailed analysis.

In addition, the COP of CRSs was analyzed according to the cascade temperature to present various research directions for improving the performance of the system. The results of this study provide a detailed analysis of CRSs according to the composition ratio of the MR. Therefore, the results are expected to be used as basic data for research on the optimization of the MR composition ratio of CRSs in consideration of both performance and exergy.

2. System Descriptions

2.1. Cascade Refrigeration System

Figure 1a is the schematic diagram of a CRS and Figure 1b shows the p-h diagram. The system consists of HTC and LTC, which are two single cycles. The cascade heat exchanger (CHX) is a heat exchanger that combines the HTC and LTC. It acts as an evaporator in the HTC and a condenser in the LTC. As can be seen from the schematic diagram, an intermediate heat exchanger (IHX) was added to the LTC. The IHX completely condenses the refrigerant of the high-pressure stream using the cooling capacity from the LTC evaporator outlet.

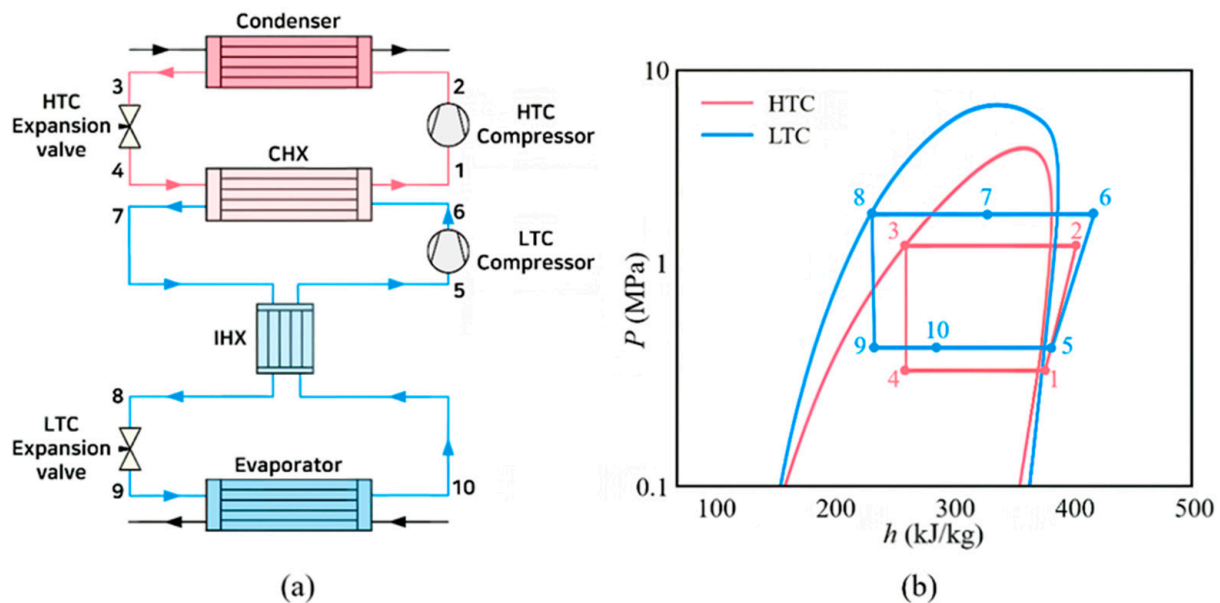


Figure 1. (a) Schematic diagram, (b) p-h diagram of the cascade refrigeration system.

Points 1 to 4 in Figure 1 represent the HTC process. The refrigerant discharged at a high temperature and high pressure from the compressor (1-2) is completely condensed through the condenser (2-3). It has a low temperature and low pressure after passing through the expansion valve (3-4) and enters CHX to exchange heat with the refrigerant of LTC (4-1). It is then introduced again to the compressor (1-2). Points 5 to 10 represent the LTC process. As with HTC, the refrigerant discharged from the compressor (5-6) exchanges heat with the refrigerant of HTC in CHX (6-7) and partially condenses. It completely condenses when passing through IHX (7-8). Subsequently, the refrigerant goes through the throttling process while passing through the expansion valve (8-9) and absorbs heat while evaporating in the evaporator (9-10). It completely evaporates while passing through IHX (10-5) and re-enters the compressor (5-6).

2.2. Selection of Refrigerants

The target temperature in this study is $-80\text{ }^{\circ}\text{C}$. A key element for clearly excluding flammability is the use of an MR composed of only non-flammable refrigerants to CRSs for the application of the MR in semiconductor processing chillers and as actual medium to large refrigeration systems. Therefore, the two factors that were considered the most important during the refrigerant selection process were the normal boiling point (NBP) and the flammability. For HTC, R-404A, a non-flammable refrigerant mainly used in previous studies on ultra-low temperature CRSs, was selected as a working fluid [6]. For LTC, the working fluid selection is very important because the composition ratio of the MR has a significant impact on the performance of the system. Additionally, the composition ratio of the MR significantly affects the energy consumption of the cryocooler [14]. Z. Zhao et al. [18] compared a two-component MR with a three-component MR and reported that the addition of a mid-boiling-point refrigerant can significantly improve the performance of the system. The application of a four-component MR to LTC improved the COP compared with the use of a three-component MR in a study on ultra-low temperature CRSs [19]. In this study, a four-component MR was applied to LTC by referring to the results of previous studies. In addition, refrigerants with different NBP were selected as working fluids to analyze the effects of high, mid, and low boiling point refrigerants on the system. In this instance, the NBP of low boiling point refrigerant is generally 40 K lower than the target temperature [23], and a refrigerant with an NBP value of approximately $-120\text{ }^{\circ}\text{C}$ is required to achieve the target temperature of $-80\text{ }^{\circ}\text{C}$. R14 was selected as the low boiling

point working fluid in this study because its NBP is $-127.8\text{ }^{\circ}\text{C}$. The physical parameters of the selected non-flammable refrigerants are shown in Table 1.

Table 1. Physical parameters of the selected refrigerants.

Refrigerants		NBP ($^{\circ}\text{C}$)	Critical Temperature ($^{\circ}\text{C}$)	Critical Pressure (MPa)	ODP	GWP	ASHRAE Safety Group
HTC	R404A	-46.2	72.04	3.73	0	3922	A1
LTC	R134a	-26.1	101.1	4.05	0	1300	A1
	R125	-48.5	66.18	3.63	0	3170	A1
	R23	-82.1	25.90	4.84	0	12,000	A1
	R14	-127.8	-45.64	3.75	0	7390	A1

3. Methodology

3.1. Assumptions and Operating Conditions

The following assumptions were made in the simulation in this study:

- (1) The system operates in a steady state;
- (2) There is no heat loss from the surroundings;
- (3) The pressure drop in the heat exchangers and pipes can be neglected;
- (4) The compression process is adiabatic, and the isentropic efficiency is a function of the compression ratio;
- (5) The evaporation and condensation processes are isobaric processes, and the expansion process is an isenthalpic process;
- (6) The cooling water that exchanges heat with the HTC condenser is supplied at $20\text{ }^{\circ}\text{C}$;
- (7) The outlet quality of the HTC condenser and LTC IHX is zero.

As mentioned in the introduction, previous studies conducted under limited pressure simulation conditions did not consider the possibility of an optimal MR composition ratio with a higher performance at the same target temperature. Therefore, in this study, the simulation was performed under various operating conditions without fixing the suction pressure. The operating parameters are listed in Table 2.

Table 2. Operating parameter values of the simulation.

Parameter	Value	Unit
Condensation temperature of HTC	30	$^{\circ}\text{C}$
Evaporation temperature of HTC	$-15\sim-40$	$^{\circ}\text{C}$
Condenser outlet quality of HTC	0	-
Temperature difference between CHX	10	$^{\circ}\text{C}$
Condensation pressure of LTC	2	MPa
Evaporation temperature of LTC	-90	$^{\circ}\text{C}$
IHX outlet quality of LTC	0	-
Cooling capacity	8	kW
Superheat degree	10	$^{\circ}\text{C}$

3.2. Thermodynamic Modeling

The thermodynamic modeling of CRSs was performed based on each point shown in Figure 1a. The equations used to analyze the energy of CRSs based on the simulation assumptions mentioned in the previous section and the operating conditions in Table 2 are shown in Table 3.

Table 3. Thermodynamic analysis equations for each component.

Component		Energy Analysis	Exergy Analysis
HTC	Compressor	$W_{Comp,H} = \frac{m_H(h_{2s}-h_1)}{\eta_{Comp,H}} = m_H(h_2 - h_1)$	$X_{Comp,H} = T_0 m_H(s_2 - s_1)$
	Condenser	$Q_{Cond,H} = m_H(h_2 - h_3)$	$X_{Cond,H} = T_0 m_H(s_3 - s_2) + Q_{Cond,H}$
	Expansion valve	$h_3 = h_4$	$X_{EV,H} = T_0 m_H(s_4 - s_3)$
CHX		$Q_{CHX} = m_H(h_1 - h_4) = m_L(h_6 - h_7)$	$X_{CHX} = T_0[m_H(s_1 - s_4) + m_L(s_7 - s_6)]$
LTC	Compressor	$W_{Comp,L} = \frac{m_L(h_{6s}-h_5)}{\eta_{Comp,L}} = m_L(h_6 - h_5)$	$X_{Comp,L} = T_0 m_L(s_6 - s_5)$
	IHX	$Q_{IHXL} = m_L(h_7 - h_8) = m_L(h_5 - h_{10})$	$X_{IHXL} = m_L T_0[(s_8 - s_7) + (s_5 - s_{10})]$
	Expansion valve	$h_7 = h_8$	$X_{EV,L} = T_0 m_L(s_9 - s_8)$
	Evaporator	$Q_{Eva,L} = Q_e$	$X_{Eva,L} = T_0[m_L(s_{10} - s_9) - \frac{Q_e}{T_{ave,Eva} + \Delta T}]$

COP(-) is a representative parameter for evaluating the thermodynamic performance of the system. It is calculated using Equation (1).

$$COP = \frac{Q_e}{W_{Comp,H} + W_{Comp,L}} \quad (1)$$

Q_e (kW) is the cooling capacity, which is fixed at 8 kW according to Table 2. $W_{Comp,H}$ (kW), $W_{Comp,L}$ (kW) are the compressor work of HTC and LTC, respectively.

Based on the simulation assumptions, the isentropic efficiency(-) of each compressor is calculated using Equation (2) [24].

$$\eta_{Comp} = 0.874 - 0.0135 \left(\frac{P_e}{P_c} \right) \quad (2)$$

P_e (MPa) is the evaporation pressure and P_c (MPa) is the condensation pressure.

The volumetric efficiency(-) of each compressor is calculated using Equation (3) [25].

$$\eta_v = 1 - C \left[\left(\frac{P_e}{P_c} \right)^{1/n} - 1 \right] \quad (3)$$

The mass flow rate of LTC (kg/s) necessary for providing a fixed cooling capacity of 8 kW is calculated using Equation (4).

$$m_L = \frac{Q_e}{h_{10} - h_9} \quad (4)$$

h_{10} (kJ/kg) and h_9 (kJ/kg) are the enthalpies at the outlet and inlet of the LTC evaporator, respectively.

CHX is a heat exchanger that combines HTC and LTC. The amount of heat transferred from CHX to HTC and LTC are the same, and they are calculated using Equation (5).

$$Q_{CHX,H} = Q_{CHX,L} = m_L(h_6 - h_7) \quad (5)$$

h_6 (kJ/kg) and h_7 (kJ/kg) are the enthalpies at the outlet and inlet of the LTC CHX, respectively.

The mass flow rate of HTC (kg/s) is calculated using Equation (6).

$$m_H = \frac{Q_{CHX}}{h_1 - h_4} \quad (6)$$

h_1 (kJ/kg) and h_4 (kJ/kg) are the enthalpies at the outlet and inlet of the HTC CHX, respectively.

The compressor displacement (m^3/s) is calculated using Equation (7),

$$V = \frac{m}{D \cdot \eta_v} \quad (7)$$

where D (kg/m^3) is the suction density of the compressor.

Exergy is a measure that represents thermodynamically useful work. Loss work and useful work can be identified through an exergy analysis of the system. The exergy destruction equations [4] used to analyze irreversible losses in each component are shown along with the energy balance equations in Table 3.

The total exergy destruction (kW) is equal to the sum of the exergy destructions in each component, which is calculated using Equation (8).

$$X_{total} = \sum X_i \quad (8)$$

3.3. Simulation Method

The simulation was performed using MATLAB software 2023. The REFPROP software 9.1 was used to import the parameters required for each part of the system, such as the temperature, pressure, and enthalpy. The simulation was performed based on the conditions in Table 2, and the mass fraction of LTC MR was changed by 0.01 units for a detailed analysis. Data reduction was then performed to refine the data obtained from the simulation. Data with temperature cross in the heat exchangers, which decreases the efficiency of the system, and data with excessive compressor displacement, which is not commercially suitable, were removed. The reliability and accuracy of the simulation were improved by identifying and removing outliers from the data. The simulation was carried out by changing the cascade temperature using the same procedure, and the specific simulation flow chart is shown in Figure 2.

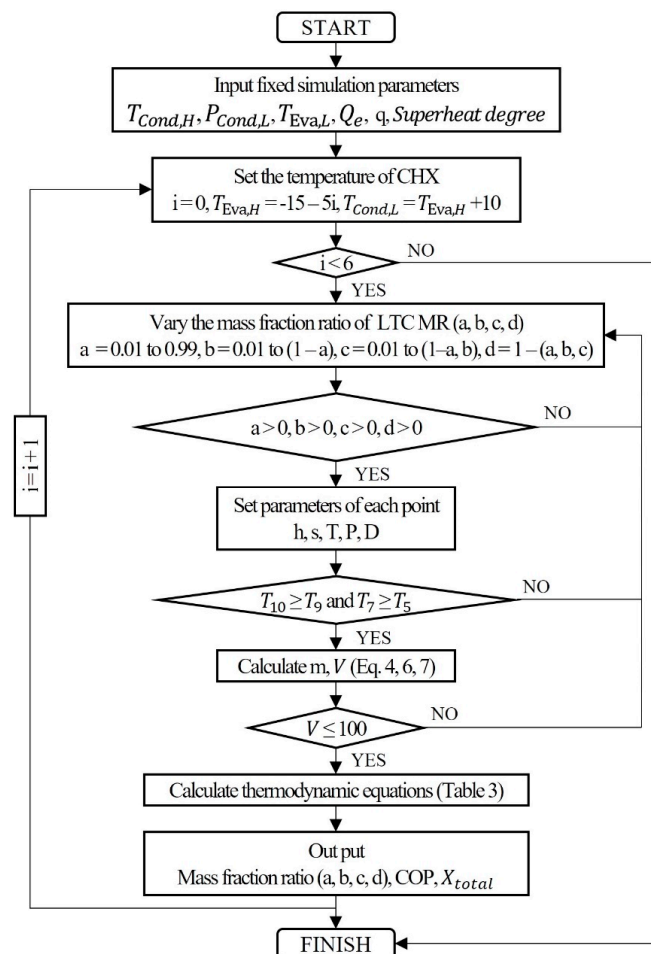


Figure 2. Simulation procedure flow chart.

4. Results and Discussion

4.1. Effect of Mass Fraction Ratio

A parametric study on the analysis of the COP and exergy of the system was conducted for a comprehensive and detailed analysis of ultra-low temperature CRSs according to the mass fraction ratio of the MR. It is possible to interpret the influence of the composition ratio of the MR in more detail by analyzing the parameters of the system that vary as four refrigerants with different boiling points increase or decrease in steps of 0.01 units. This will be helpful in understanding the effects of the proportions of high, mid, and low boiling point refrigerants on the operation of the system and in finding the optimal conditions.

4.1.1. Coefficient of Performance

Figure 3 shows the COP change according to the mass fraction of the MR for an evaporation temperature of HTC of $-15\text{ }^{\circ}\text{C}$. The highest COP was 0.51 when the mass fractions of R134a, R125, R23, and R14 were 0.15, 0.2, 0.11, and 0.54, respectively. When the mass fraction of R134a remained constant at 0.15, COP increased as the mass fraction of R125 increased. As the mass fraction of R23 increased, COP went through a rising section, a sharp decreasing section, and a gradual rising section in most cases.

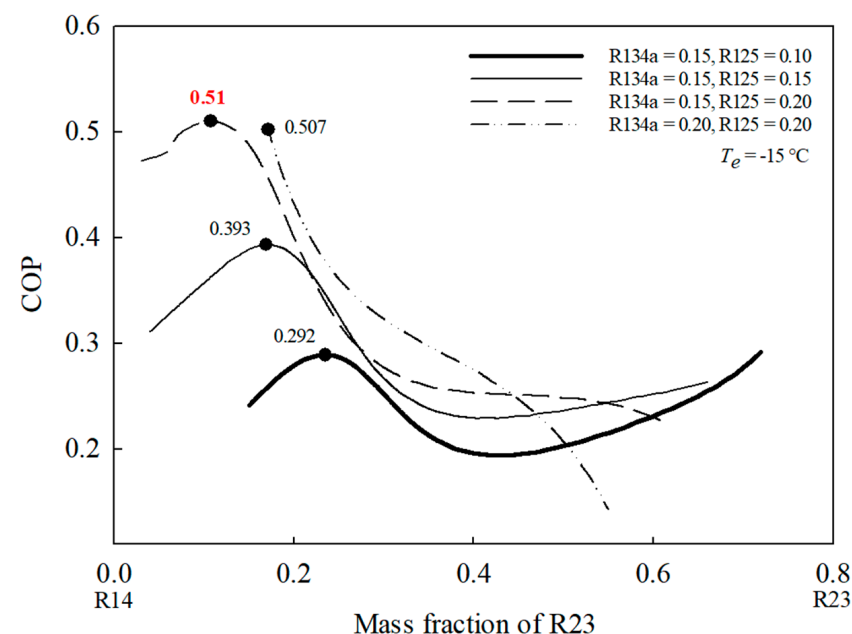


Figure 3. Variation of COP according to mass fraction ratio.

COP was calculated using Equation (1), and the cooling capacity was fixed at 8 kW under the simulation conditions shown in Table 2. Therefore, it was necessary to analyze the compressor work acting as a variable when analyzing the unique COP trend according to the mass fraction of the MR, as shown in Figure 4.

In Figures 4 and 5, the black line is used to distinguish the thickness of the red and blue lines representing HTC and LTC, respectively. It can be shown that the impact of the LTC is dominant in the compressor work even though HTC and LTC showed the same trend with regard to the increase and decrease in the compressor work. Figure 5 shows the mass flow rates of HTC and LTC under the same conditions. As with Figure 4, the impact of HTC was not significant, and the mass flow rate of LTC tended to decrease continuously as the mass fraction of R23 increased. In addition, the mass flow rate decreased as the mass fraction of high boiling point refrigerants (R134a and R125) increased. The mass flow rate is inversely proportional to the enthalpy difference between the inlet and outlet of the LTC evaporator, i.e., the refrigeration effect, according to Equation (4). Figure 6 shows the change in refrigeration effect. As the proportion of the high boiling point refrigerants increased, the refrigeration effect increased, thereby reducing the mass flow rate. The

refrigeration effect (Joule-Thomson coefficient) of the MR is high when the proportion of the high boiling point refrigerants is high and that of the low boiling point refrigerants is low [26].

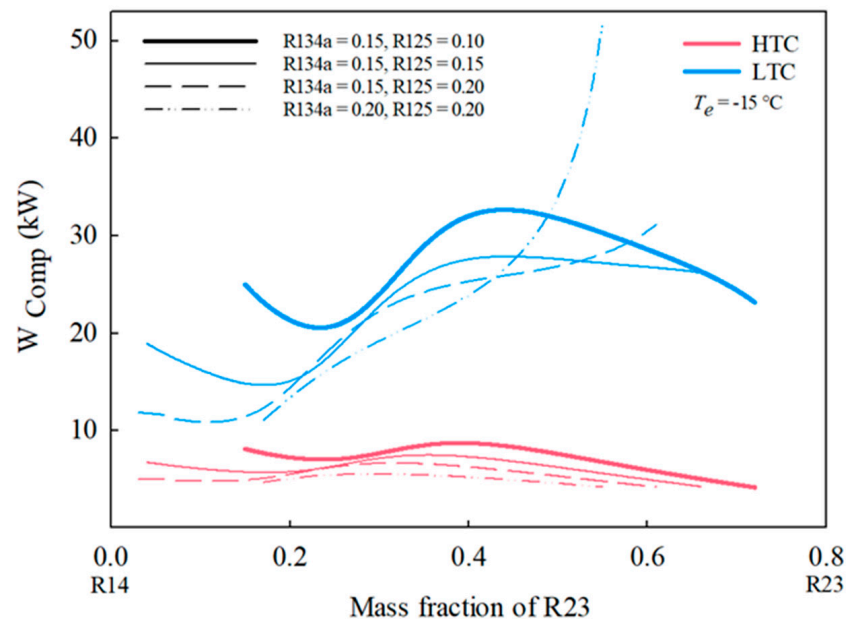


Figure 4. Variation of compressor work according to mass fraction ratio.

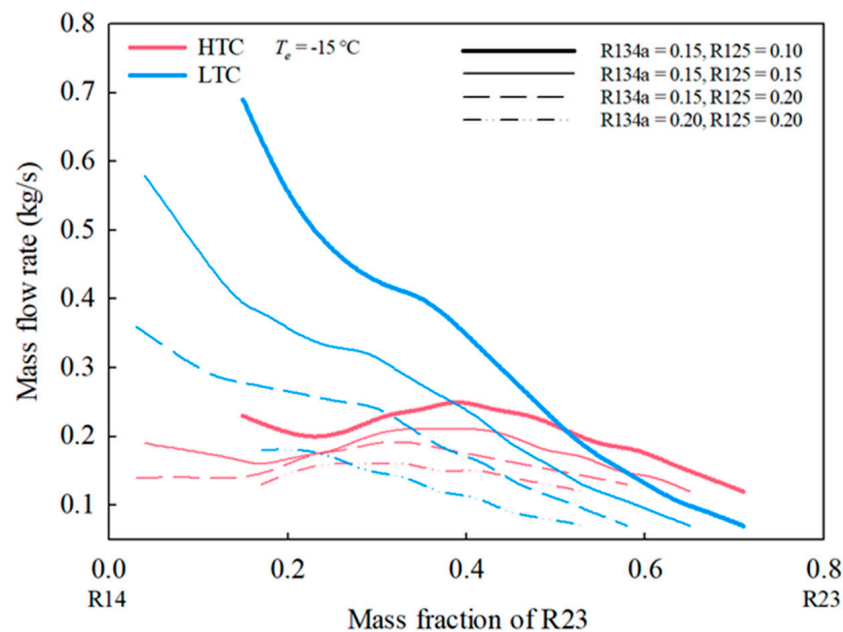


Figure 5. Variation of mass flow rate according to mass fraction ratio.

It should be noted in Figure 6, however, that the highest COP point in Figure 3 appeared in the section in which the mass fraction of R23 with a relatively low refrigeration effect was lower than 0.3. In other words, it cannot be concluded that the performance of the system is low just because the refrigeration effect is low, and the results show the limitations of using only the refrigeration effect as a performance indicator for MR selection for ultra-low temperature CRSs. To analyze the cause of this trend, the LTC isentropic efficiency and the enthalpy difference of the LTC compressor, which are parameters affecting the LTC compressor work according to Figure 4, are shown in Figures 7 and 8, respectively.

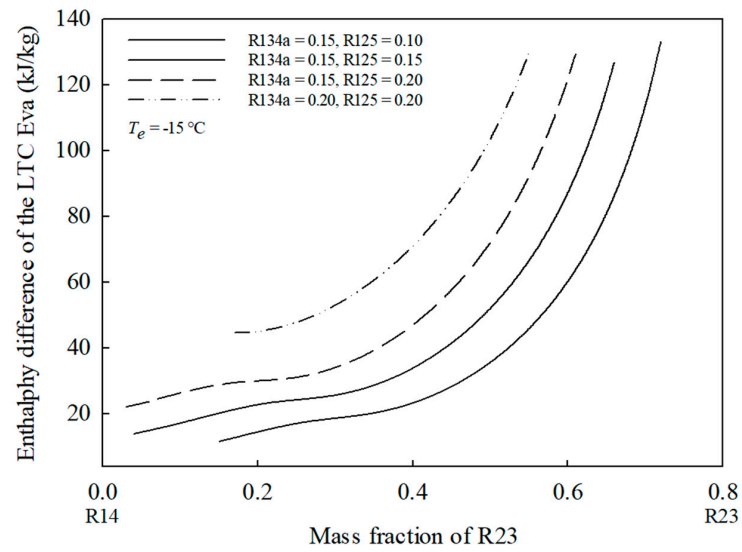


Figure 6. Variation of enthalpy difference of the LTC evaporator according to mass fraction ratio.

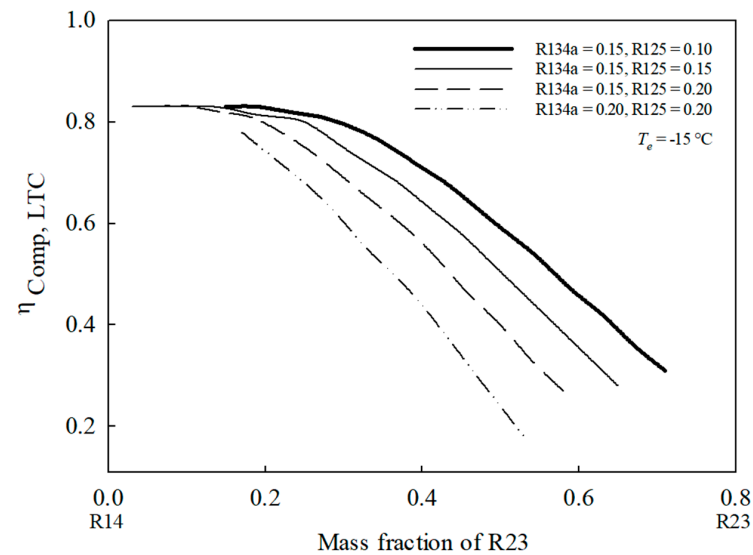


Figure 7. Variation of LTC compressor isentropic efficiency according to mass fraction ratio.

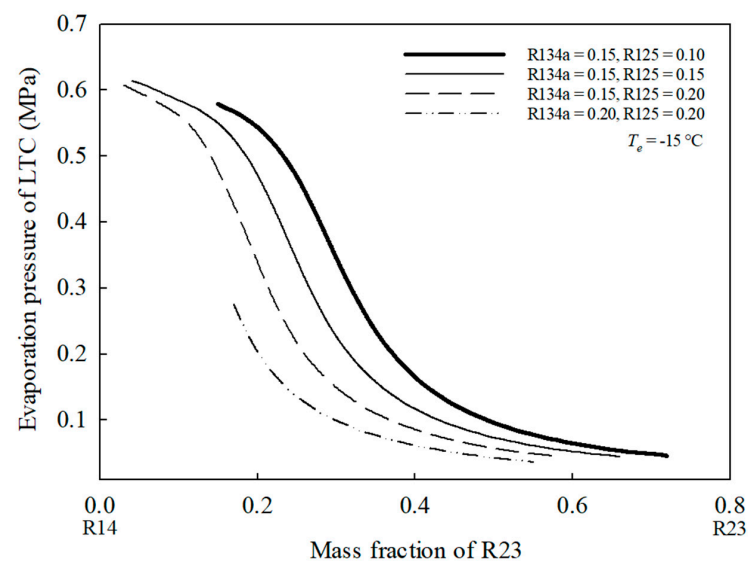


Figure 8. Variation of LTC evaporation pressure according to mass fraction ratio.

As can be seen in Figure 7, the compression efficiency decreased as the mass fraction of the high boiling point refrigerants increased. This is because the compression efficiency calculated using Equation (2) was affected by the reduction in evaporation pressure shown in Figure 8. When the quantities of the low boiling point components are insufficient, the evaporation pressure and mass flow rate decrease [14].

Figure 9 shows the enthalpy difference between the inlet and outlet of the LTC compressor. The enthalpy difference tended to increase as the proportion of refrigerants with a relatively high boiling point increased.

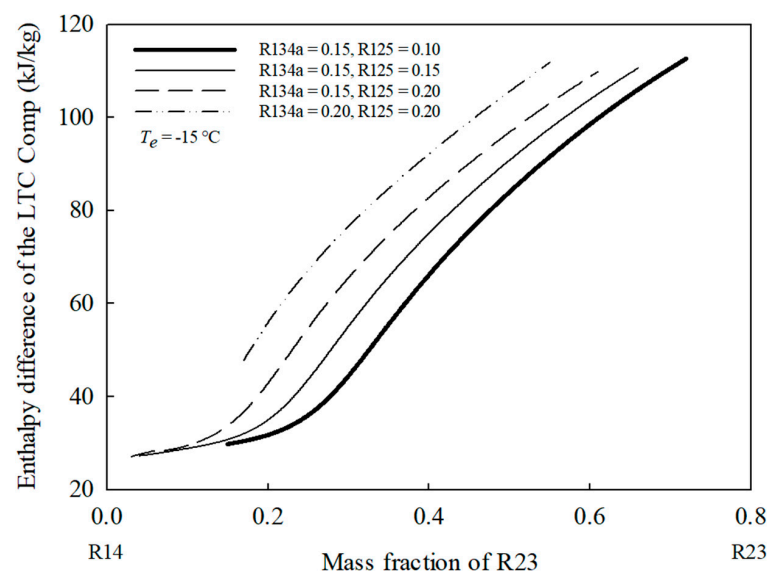


Figure 9. Variation of enthalpy difference of the LTC compressor according to mass fraction ratio.

In summary, as the proportion of refrigerants with relatively high boiling points increased, the mass flow rate of LTC decreased, the enthalpy difference of the compressor increased, and the isentropic efficiency decreased. These are parameters that directly affect the compressor work, meaning that the compressor work was divided into decreasing, increasing, and decreasing again sections instead of a continuous increase or decrease as in Figure 4, due to the presence of a dominant parameter for each section according to the mass fraction of MR.

In the stage in which the compressor work of LTC decreased, the change in isentropic efficiency was insignificant (up to 5.13%). In addition, the mass flow rate decreased by up to 27.54% and the enthalpy difference of the compressor increased by up to 17.56%. In this section, in which the mass fraction of R23 was relatively low (less than 0.23), and that of R14 was higher than 0.5, the influence of the reduction in the mass flow rate was dominant. In other words, when the proportion of the low boiling point refrigerant was more than half of the MR, the mass flow rate decreased rapidly. Despite an increase in the compressor enthalpy difference, the compressor work decreased and COP increased, as shown in Figure 3.

In the stage in which the compressor work rapidly increased, the slope of the reduction in the mass flow rate decreased, indicating that this stage was significantly affected by the increase in compressor enthalpy difference and the decrease in compression efficiency.

In the stage in which the compressor work decreased again, the compressor enthalpy difference increased and the compression efficiency decreased, but the influence of the reduction in the mass flow rate was dominant.

When the trends of the parameters according to the mass fraction of the MR were analyzed in detail, it was found that the COP increased as the proportion of the high boiling point refrigerants increased. Regardless of the proportion of the high boiling point refrigerants, as the mass fraction of R23 increased, a low boiling point refrigerant (R14) was further squeezed, which resulted in a decrease in COP. It was mentioned that the lack of a

low boiling point working fluid is the main cause of reducing the COP of an MR chiller at the low target temperature [19]. In other words, it is desirable for the mass fraction of the low boiling point refrigerant to exceed 0.5 in ultra-low temperature CRSs targeted at $-80\text{ }^{\circ}\text{C}$. When the mass fraction of the mid boiling point refrigerant was approximately 0.3, a high COP was observed. However, it is important to pay careful attention because the COP decreases if the mass fraction of the low boiling point refrigerant is excessively high, as shown in Figure 3. It has been experimentally revealed that the cooling capacity and cooling performance decrease when the proportion of the low boiling point refrigerants is excessively high in a JT cooler with a target temperature of $-60\text{ }^{\circ}\text{C}$ [14]. Therefore, a mass fraction of between 0.5 and 0.55 for the low boiling point refrigerants is favorable for COP improvement; this is neither insufficient nor excessive.

4.1.2. Exergy Analysis

In this section, the mass fraction ratio of MR, which can improve efficiency by identifying the irrationality of the energy utilization process, was analyzed through exergy analysis. In addition, the irreversibility positions of each component of the cycle were shown to present potential thermodynamics improvement directions.

Figure 10 shows the total exergy destruction according to the mass fraction of MR for an evaporation temperature for HTC of $-15\text{ }^{\circ}\text{C}$. The highest total exergy destruction was 35.24 kW when the mass fractions of R134a, R125, R23, and R14 were 0.15, 0.1, 0.41, and 0.34, respectively. The lowest total exergy destruction was 11.58 kW when the mass fractions were 0.15, 0.2, 0.1, and 0.55, respectively. The total exergy destruction tended to decrease when the mass fraction of R23 was lower than 0.23 and when it was higher than 0.4. In addition, when the mass fraction of R134a was fixed at 0.15, the total exergy destruction tended to decrease as the mass fraction of R125 increased. In other words, the total exergy destruction decreased as the mass fraction of the high boiling point refrigerant increased and that of the low boiling point refrigerant decreased.

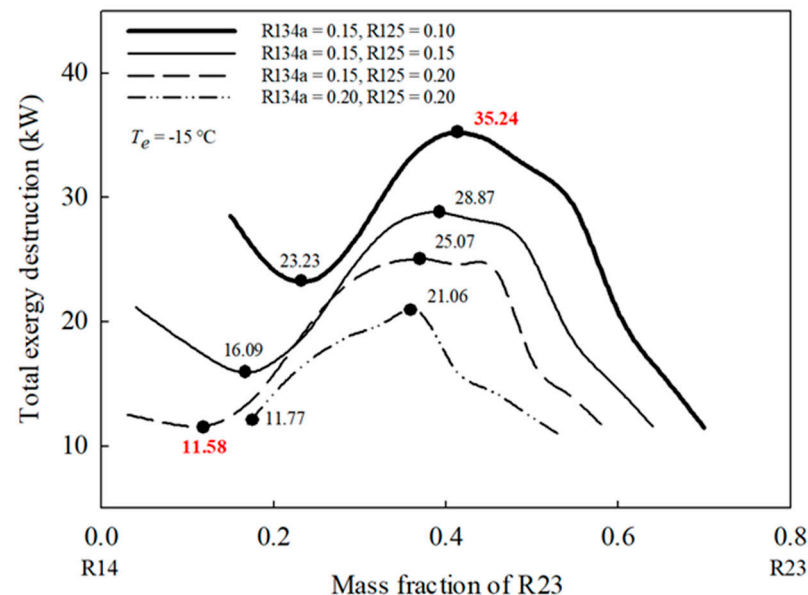


Figure 10. Variation of total exergy destruction according to mass fraction ratio.

Figure 11 shows the discharge temperature of the LTC compressor according to the mass fraction ratio of MR. It can be seen that the discharge temperature tended to increase as the mass fraction of the high boiling point refrigerants increased. When the mass fraction of R23 was fixed at 0.4, the discharge temperature for R134a and R125 mass fractions of 0.2 and 0.2, respectively, was 35.09% higher than that for R134a and R125 mass fractions of 0.15 and 0.1, respectively. In addition, the compressor discharge temperature increased as the mass fraction of R23 increased, and the discharge temperature exceeded $100\text{ }^{\circ}\text{C}$ when

the mass fraction of R14, a low boiling point refrigerant, was less than 0.1. The compressor discharge temperature is a very important factor in the system design process, and a high discharge temperature may have a negative impact on the refrigerant and lubricant, as it increases the risk of equipment shutdown and adversely affects the performance and service life of the entire system, including the compressor. It is, therefore, very important to improve the stability and efficiency of the system by properly managing the discharge temperature. Therefore, considering the discharge temperature rise in Figure 11, it can be said that the first reduction section of the total exergy destruction in Figure 10, for which the mass fraction of R23 was less than 0.23, is the optimal point.

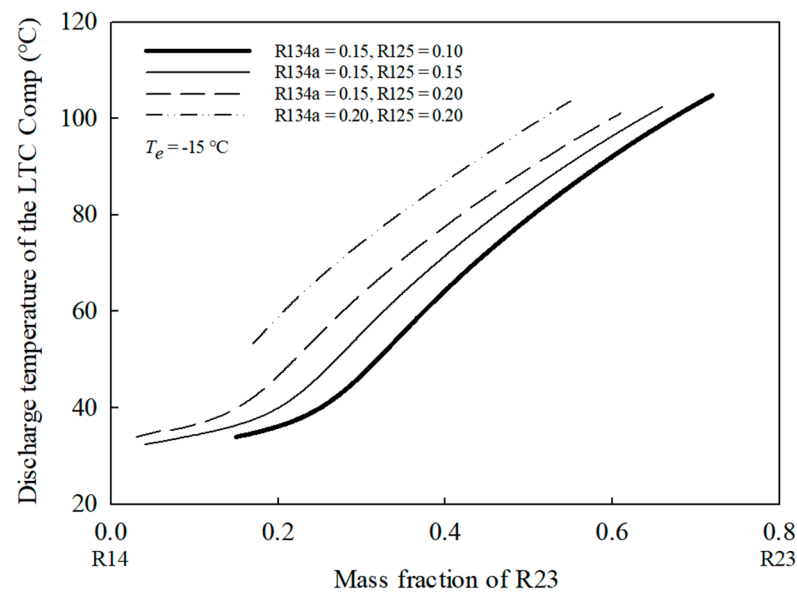


Figure 11. Variation in discharge temperature of the LTC compressor according to mass fraction ratio.

Figure 12 shows the COP and total exergy destruction of the system according to the mass fraction ratio considering the efficiency of the system in terms of both energy and exergy. It was found that the COP is inversely proportional to the total exergy destruction. When everything, i.e., the LTC compressor discharge temperature, total exergy destruction, and COP, were considered, the ideal mass fractions of R134a and R125 ranged between 0.1 and 0.2, respectively, and a mass fraction for R23 of less than 0.23 was found to be the optimal solution. To more accurately evaluate the degree of irreversibility of the cycle within this optimal mass fraction ratio section, an exergy destruction analysis was conducted for all components.

For the analysis, the composition ratio of MR within the optimal mass fraction ratio range was fixed for four cases, as shown in Table 4.

Figure 13 shows the exergy destruction rate by component for the four cases in Table 4 for an evaporation temperature of the HTC of $-15\text{ }^{\circ}\text{C}$. In all cases, the exergy destruction rate of LTC was higher than that of HTC. This indicates that an intensive analysis of LTC is required to improve the efficiency of the energy utilization process in CRSs.

Table 4. Mass fraction case number for MR in the optimal range.

Case Number	Mass Fraction			
	R134a	R125	R23	R14
1	0.15	0.10	0.23	0.52
2	0.15	0.15	0.2	0.5
3	0.15	0.20	0.16	0.49
4	0.20	0.20	0.17	0.43

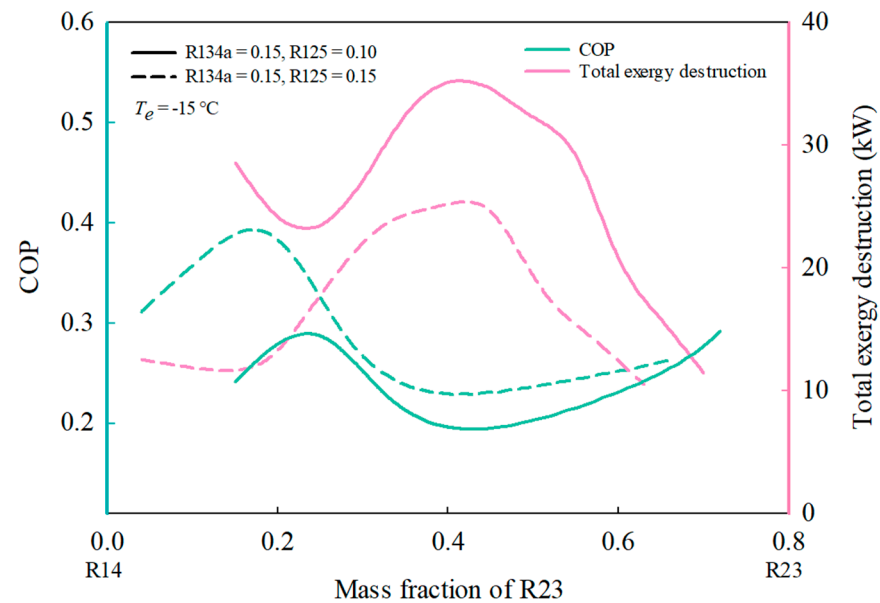


Figure 12. Variation of COP and total exergy destruction according to mass fraction ratio.

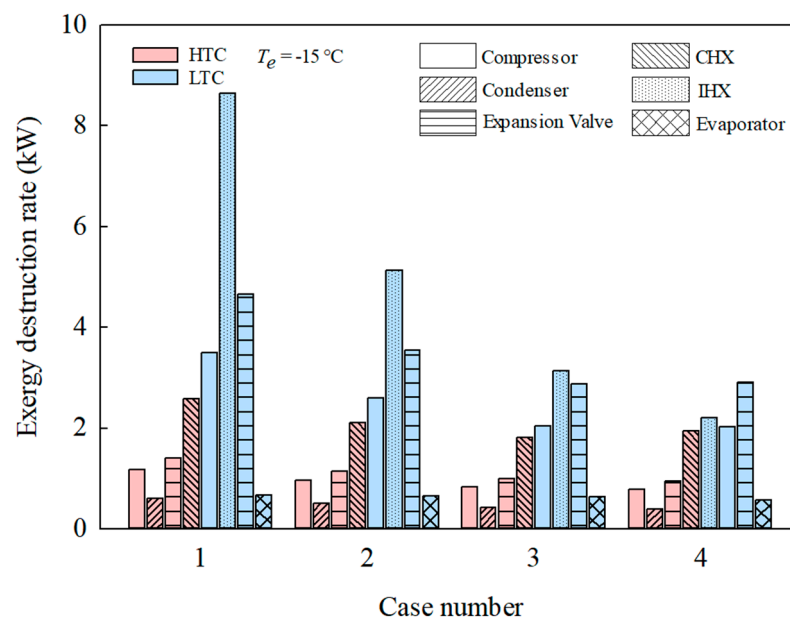


Figure 13. Exergy destruction rate by component according to different mass fraction case.

In case 1, the exergy destruction rate of IHX was 8.65 kW, which was the highest among the exergy destruction rates by component of all cases. In most cases, IHX exhibited the highest exergy destruction rate. In case 1, the mass fractions of R134a and R125 were 0.1 and 0.15, respectively. The proportion of the high boiling point refrigerants was the lowest among all cases. As can be seen from Figure 5, the mass flow rate of LTC increased as the proportion of the high boiling point refrigerants decreased. The relatively high mass flow rate in case 1 caused IHX to require more resilience, resulting in the highest exergy destruction rate. As a result of comparing and analyzing the exergy destruction of MR according to the temperature difference of the heat exchanger, it is possible to reduce irreversible losses and improve the performance of the system by decreasing the heat exchanger temperature difference [23]. If the system is designed to reduce the temperature difference between the high-temperature fluid and the low-temperature fluid in the IHX by increasing the heat exchange area, it helps to reduce the exergy destruction in the IHX.

Apart from in the IHX, large irreversible losses occurred in the expansion valve and compressor of the LTC. In cases 1, 2, 3, and 4, the exergy destruction rates of the expansion valve were 4.65, 3.55, 2.88, and 2.90 kW, and those of the compressor were 3.49, 2.59, 2.05, and 2.21 kW, respectively. As with IHX, the largest exergy destruction occurred in case 1, for which the amounts of the high boiling point refrigerants were relatively low. This appears to be due to an increase in the mass flow rate, as described above. Therefore, the exergy destruction rates of the expansion valve and compressor can be decreased by increasing the mass fraction of the high boiling point refrigerants in the MR, thereby reducing the mass flow rate.

4.2. Effect of Cascade Temperature

A CRS is a system in which numerous parameters, such as temperature and pressure, interact in the evaporation and condensation processes of two single cycles according to the temperature of CHX. Figure 14 shows (a) COP and (b) COP of the HTC and LTC according to the evaporation temperature of the HTC from previous studies on a CRS [27–32]. When a single refrigerant is applied in a CRS, the HTC COP of the CRS increases, and the COP of LTC decreases as the cascade temperature increases, as shown in Figure 14b. Finally, COP tends to increase and then decrease according to the cascade temperature, as shown in Figure 14a. Thus, the COP trend according to the cascade temperature was analyzed additionally when MR was applied to the CRS to present further research directions for system performance improvement through a comparison with previous studies. The MR mass fraction for the analysis was fixed, as in Table 5.

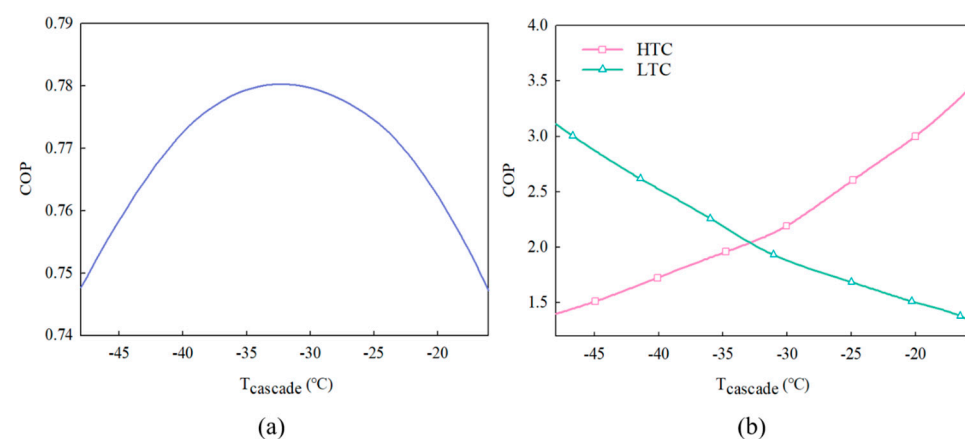


Figure 14. (a) Variation of COP, (b) Variation COP of HTC and LTC according to cascade temperature in previous literature.

Table 5. MR case number for analyzing COP according to cascade temperature.

MR Number	Mass Fraction			
	R134a	R125	R23	R14
MR 1	0.05	0.20	0.23	0.52
MR 2	0.08	0.12	0.28	0.52

Figure 15 shows the COP tendencies of MR1 and MR2 according to the evaporation temperature of HTC. Since MR1 and MR2 have different mass fractions, the COPs do not align, as shown in Figure 15. However, the similar trends in COP with varying cascade temperatures effectively illustrate the influence of cascade temperature on COP. For both MR1 and MR2, COP tended to decrease as the cascade temperature increased. To analyze the cause of this tendency, the compressor work of HTC and LTC is shown in Figure 16 according to the cascade temperature.

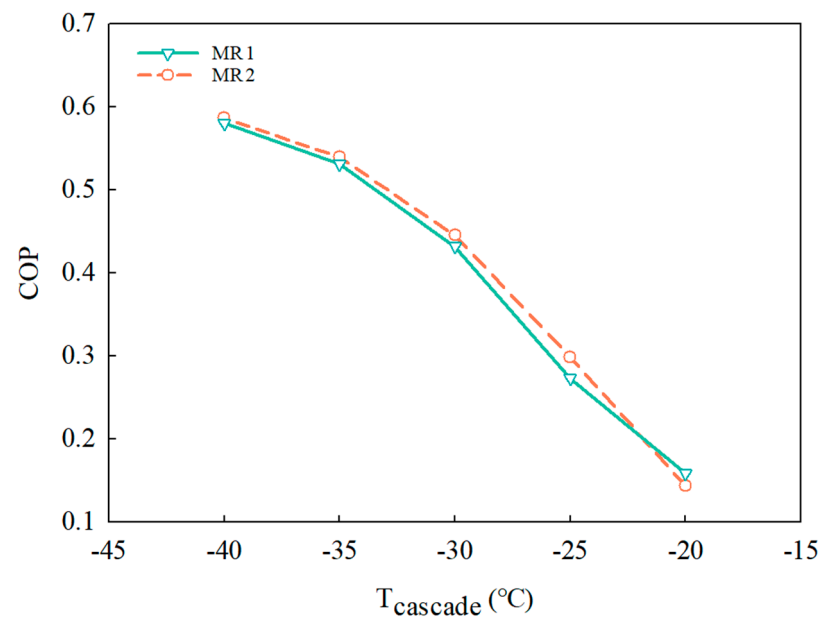


Figure 15. Variation in COP according to cascade temperature.

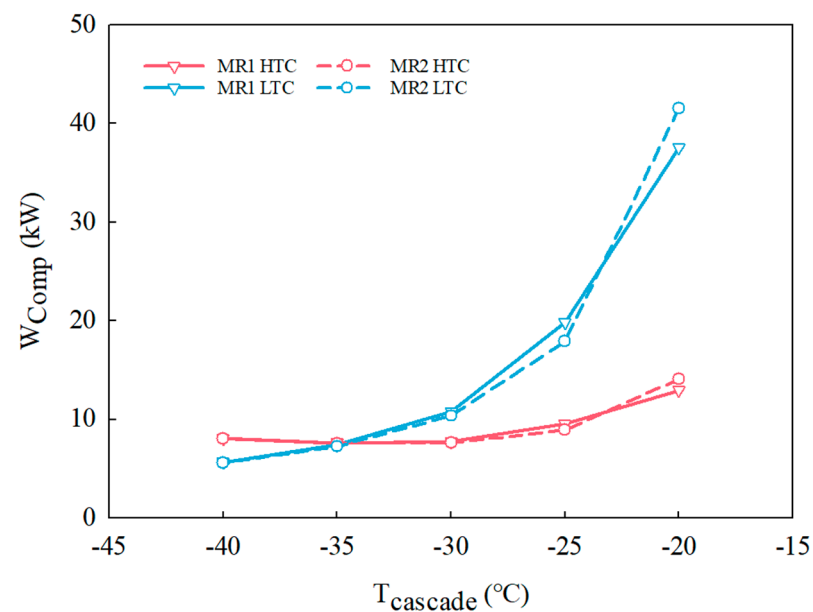


Figure 16. Variation in compressor work according to cascade temperature.

As shown in Figure 16, the compressor work of LTC continuously increased as the cascade temperature increased. The compressor work of HTC decreased in the range of -40 to -30 $^{\circ}\text{C}$ and then tended to increase above -30 $^{\circ}\text{C}$. In the -40 to -30 $^{\circ}\text{C}$ temperature range, the LTC compressor work increased by 89.56% and 85.61% for MR1 and MR2, respectively, while the HTC work decreased by 10.8% and 5.59%, respectively. The increase rate of the LTC compressor work was much higher than the decrease rate of the HTC compressor work. Thus, COP decreased, as shown in Figure 15. Above -30 $^{\circ}\text{C}$, the compressor work of both HTC and LTC increased. Therefore, COP continuously decreased as the cascade temperature increased. It is well known that an increase in the cascade temperature leads to an increase in the compression ratio and compression work for LTC. For HTC, however, the compression work tended to increase above a certain temperature as the cascade temperature increased rather than continuously decreasing as the compression ratio decreased. Therefore, the HTC mass flow rate, a parameter that affects the HTC compressor work, was analyzed. The HTC mass flow rate is calculated using Equation (6).

As it is proportional to the heat load of CHX, the HTC mass flow rate and the heat load of CHX for MR1 according to the cascade temperature are shown in Figure 17.

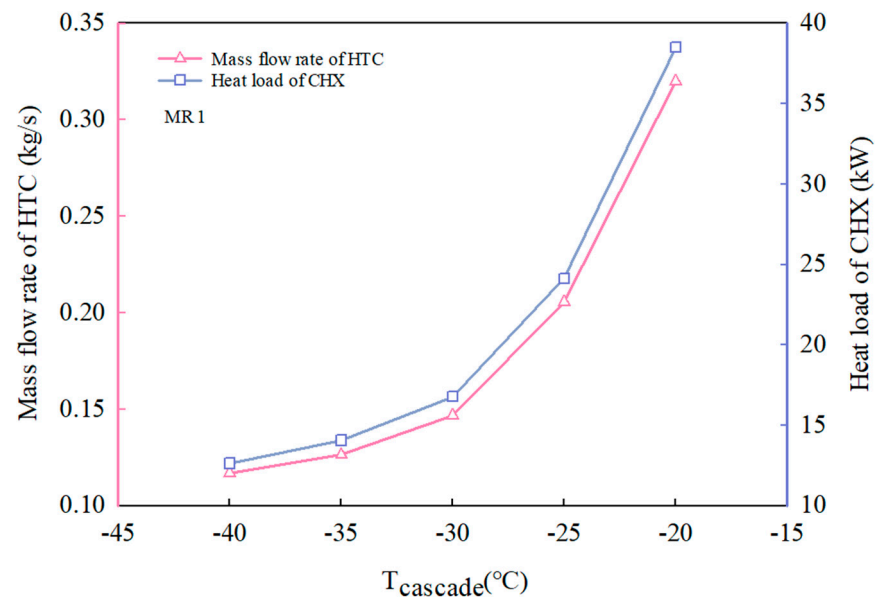


Figure 17. Variation in the mass flow rate of HTC and the heat load of CHX according to cascade temperature.

As shown in Figure 17, the mass flow rate of HTC continuously increased as the cascade temperature increased. The increase rate of the mass flow rate of HTC was 25.48% in the -40 to -30 $^{\circ}\text{C}$ temperature range, while it was 118.24% in the -30 to -15 $^{\circ}\text{C}$ temperature range. This caused the HTC compressor work to increase according to Equation (5), under the influence of the increase in the mass flow rate rather than continuously decreasing, as in Figure 16. Consequently, COP tended to decrease as the cascade temperature increased, as can be seen in Figure 15, showing a completely different trend from the results in previous studies that applied a single refrigerant, as shown in Figure 14. Therefore, when a system is designed by applying an MR to a CRS, it is necessary to properly select the cascade temperature for COP improvement.

4.3. Comparison: Conventional vs. Proposed Method

This section provides a comparative analysis of simulation methods utilized in prior research and the methodology proposed in this study. The objective is to highlight the advancements in energy efficiency and exergy performance that the proposed method offers, in contrast to earlier approaches where high and low pressures were fixed. Simulation for the conventional method was conducted based on the conditions presented in Table 6, with the results depicted in Figures 18 and 19.

Table 6. Operating parameter values of the conventional method simulation.

Parameter	Value	Unit
Condensation temperature of HTC	30	$^{\circ}\text{C}$
Evaporation temperature of HTC	$-15 \sim -40$	$^{\circ}\text{C}$
Condenser outlet quality of HTC	0	-
Temperature difference between CHX	10	$^{\circ}\text{C}$
Condensation pressure of LTC	2	MPa
Evaporation temperature of LTC	-90	$^{\circ}\text{C}$
Evaporation pressure of LTC	0.2	MPa
Cooling capacity	8	kW
Superheat degree	10	$^{\circ}\text{C}$

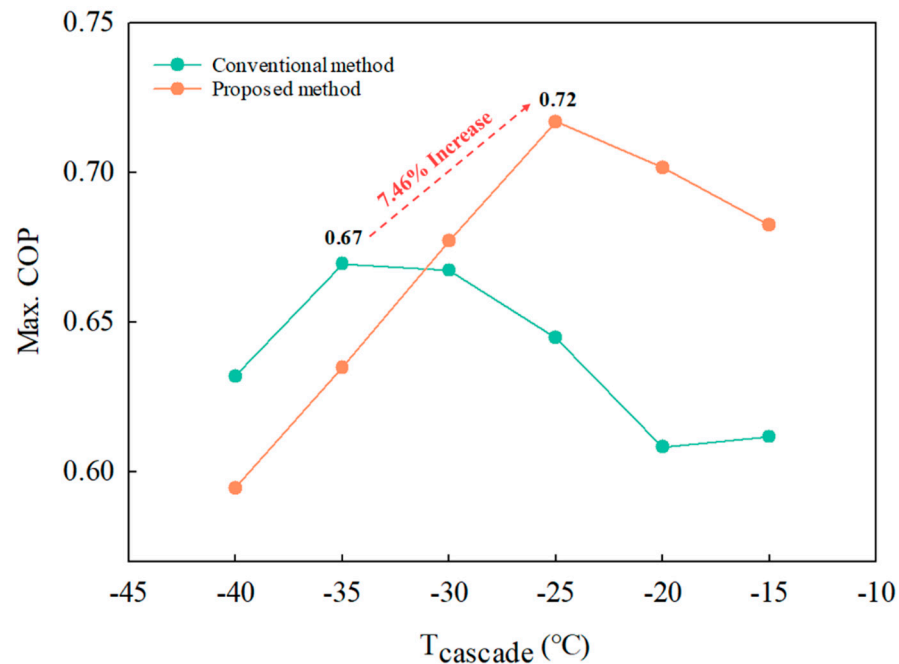


Figure 18. Comparison of conventional and proposed method maximum COP according to cascade temperature.

Figure 18 shows the maximum COP values for both the conventional and proposed methods according to the evaporation temperature of HTC. When the evaporation temperature of HTC is above -30°C , the maximum COP is consistently higher with the proposed method compared to the conventional method. Furthermore, for temperatures below -35°C , the conventional method shows a higher maximum COP. However, the proposed method achieves a maximum COP of 0.72 at -25°C , representing a 7.46% improvement over the conventional method's maximum COP of 0.67 at -35°C . This suggests that the proposed method, by not fixing the suction pressure, is capable of identifying MR composition with superior COP, highlighting a limitation in previous research that did not account for potentially more efficient MR composition for the same target temperature.

Figure 19 compares the total exergy destruction for the MR composition with the maximum COP shown in Figure 18, according to the evaporation temperature of HTC for both methods. In Figure 18, the temperature corresponding to the maximum COP for the proposed and conventional methods are -25°C and -35°C , respectively. Comparing the total exergy destruction at these specific temperatures is crucial. Evaluating exergy destruction at these maximum COP points enables an accurate assessment of the proposed method's efficiency and effectiveness relative to the conventional method under equivalent performance conditions. At -25°C , where the proposed method achieves the maximum COP, the total exergy destruction is 7.13 kW. In contrast, at -35°C , where the conventional method achieves its maximum COP, the total exergy destruction is 7.58 kW. The proposed method thus results in a 5.94% reduction in exergy destruction compared to the conventional method. This indicates that the proposed method offers an MR composition with both a higher maximum COP and lower exergy destruction simultaneously.

Figures 18 and 19 demonstrate that the proposed method provides an MR composition with superior energy and exergy performance compared to those obtained using the conventional method. This finding is particularly relevant for ultra-low temperature processes, such as semiconductor applications, where optimizing MR composition can significantly enhance the efficiency of ultra-low temperature chillers.

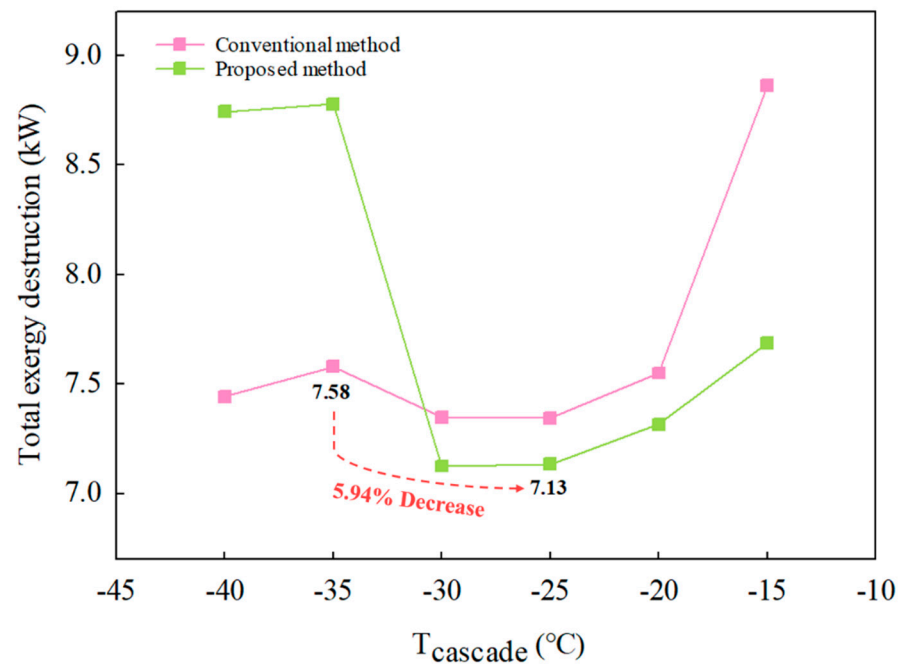


Figure 19. Comparison of conventional and proposed method total exergy destruction according to cascade temperature.

5. Conclusions

In this study, the effects of the mass fractions of high, mid, and low boiling point refrigerants on the system are discussed, and the optimal conditions in consideration of the coefficient of performance (COP) and the exergy are presented through a comprehensive and detailed analysis of an ultra-low temperature cascade refrigeration system (CRS) according to the mass fraction ratio of the mixed refrigerant (MR). Based on the simulation results, the following conclusions were drawn:

- (1) The simulation was performed by varying the MR in steps of 0.01 units without fixing the suction pressure to optimize the composition ratio under various operating conditions.
- (2) The COP initially increased, then decreased, and increased again as the mass fraction of R23 increased. The highest COP was observed when the refrigeration effect was low (R134a, R125, R23, and R14: 0.15, 0.2, 0.11, and 0.54), suggesting that using refrigeration effect alone as a performance indicator has limitations in selecting an MR for ultra-low temperature CRSs.
- (3) A mass fraction of low boiling point refrigerants between 0.5 and 0.55 improved COP, while COP was inversely proportional to total exergy destruction.
- (4) The optimal mass fractions for COP and exergy destruction occurred with R134a and R125 between 0.1 and 0.2 and R23 less than 0.23. The IHX in the LTC showed the highest exergy destruction rate, and reducing the temperature discrepancy and mass flow rate could reduce exergy destruction.
- (5) As the cascade temperature increased, COP decreased, emphasizing the importance of selecting the optimal cascade temperature for performance improvement. The proposed method showed a 7.46% improvement in maximum COP and a 2.99% reduction in total exergy destruction compared to the conventional method.

This study provides valuable insights into optimizing the composition ratio of MR for ultra-low temperature CRSs. It will serve as a foundation for future experimental research, which will include validating the simulation results to further enhance the findings. The findings highlight the necessity of considering a broader range of operational parameters beyond traditional metrics like refrigeration effect and existing simulation methods. The optimal conditions revealed provide a practical approach to enhancing performance and

efficiency in semiconductor processing chillers and similar applications. Further research should explore additional refinements and practical implementations of these findings to advance the development of more efficient and sustainable refrigeration technologies.

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Conflicts of Interest: The author declares no conflict of interest.

Nomenclature

SYMBOLS

D	Density	kg/m^3
h	Enthalpy	kJ/kg
m	Mass flow rate	kg/s
P	Pressure	MPa
q	Quality	
Q	Heating or cooling capacity	kW
s	Entropy	$\text{kJ}/\text{kg}\cdot\text{K}$
T	Temperature	$^{\circ}\text{C}$
V	Compressor displacement	m^3/s
W	Power	kW
X	Exergy destruction	kW

SUBSCRIPTS

0	Ambient
1-10	State point
ave	Average
c	Condensation
cascade	Cascade heat exchanger
Comp	Compression or compression
Cond	Condenser or condensation
e	Evaporation
Eva	Evaporator
EV	Expansion valve
H	High-temperature cycle
i	System components
L	Low-temperature cycle
s	Isentropic
total	Total system components

GREEK

SYMBOLS

η_{Comp}	Isentropic efficiency of compressor
η_v	Volumetric efficiency of compressor
Δ	Difference

ABBREVIATIONS

AI	Artificial intelligence
CHX	Cascade heat exchanger

COP	Coefficient of performance
CRS	Cascade refrigeration system
GWP	Global warming potential
HFO	Hydrofluoroolefin
HTC	High-temperature cycle
IHX	Intermediate heat exchanger
JT	Joule-Thomson
LTC	Low-temperature cycle
MR	Mixed refrigerant
NBP	Normal boiling point (°C)
ODP	Ozone depletion potential

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