### Articles



## Utilizing of Mixture Refrigerant in Brayton Cycle Using Turbomachinery

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TAIYO NIPPON SANSO corporation uses various refrigeration cycles in the production and application of industrial gases such as oxygen, nitrogen and argon. One of them is the Brayton cycle that uses adiabatic compression and adiabatic expansion with turbomachinery (turbo-Brayton cycle). In 2013, we commercialized a turbo-Brayton cycle refrigerator that uses neon as a refrigerant for cooling superconductivity power equipment. The refrigerant of the turbo-Brayton cycle does not liquefy at the cooling temperature, and the design of turbomachinery must be established according to the physical properties. Therefore, neon was selected for our commercialized refrigerator. Also, the pressure ratio of the turbo-Brayton cycle is chosen for high efficiency. On the other hand, depending on the pressure ratio and the temperature condition of the fluid to be cooled, the fluid to be cooled may be below the freezing point, so measures to prevent freezing are taken.

Recent studies have shown that the utilizing of mixture refrigerant provides a wider range for pressure ratio and turbomachinery design, and solves the problem of solidification of the fluid to be cooled while keeping up performance of the refrigerator. In this paper, we report the concept, advantages and the result of specific study of utilizing mixture refrigerant in the turbo-Brayton cycle.

### 1. Introduction

In the early 1900s, equipment for liquefying air and separating oxygen (air separation unit) was developed, and the technology for industrial handling of low-temperatures below 110 K was established. The cold generation in air separation units initially used Joule-Thomson expansion valves, followed by reciprocating expanders, and then by turbine-type expanders (expansion turbines), which are compact and capable of regulating flow with low losses.

On the other hand, in the field of cryogenic research, the realization of a refrigeration cycle to liquefy helium (in the temperature range of 4 K) led to the discovery of superconductivity (a phenomenon in which electrical resistance is reduced to zero). In the 1950s, expansion turbines were also used in helium liquefiers to improve the liquefying capacity, which encouraged the development of research in the fields of cryogenic and superconductivity <sup>1</sup>).

Thus, advances in the refrigeration cycle necessary for lowtemperature generation and in the technology of turbomachinery, such as expansion turbines used in the refrigeration cycle, have played an important role in the development of industrial fields such as air separation units and helium liquefiers, as well as of the research fields such as cryogenics and superconductivity.

In the 1980s, copper oxide superconductors (hightemperature superconductors) such as yttrium-based and bismuth-based superconductors were discovered, and a breakthrough occurred in which the temperature at which the transition to superconductivity occurs (critical temperature) changed from the 4 K range to the 77 K range. Since liquid helium is not required for cooling high-temperature superconductors, cooling energy and cooling costs can be reduced, which accelerated the research on application of high-temperature superconductors to superconductivity cables, superconductivity motors, and other power equipment. As a result, empirical research on practical-scale superconductivity power equipment progressed in the 2000s, and it became essential to make practical a refrigerator with a cooling capacity and long maintenance intervals suitable therefor. To meet these requirements, in 2013 we developed

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a turbo-Brayton cycle refrigerator using a turbomachinery that cools sub-cooled liquid nitrogen <sup>2</sup>). This technology was also adopted for the project of the world's first commercial superconductivity cable in Korea <sup>3</sup>), which is still operating successfully.

Recently, as part of efforts toward a decarbonized society, the use of hydrogen as an energy source has been actively studied, which includes the use of liquid hydrogen (20 K) <sup>4</sup>). The application of the turbo-Brayton cycle to pre-cooling of the liquefying system required to liquefy hydrogen is being considered in such efforts <sup>5</sup>) as well.

Thus, the required temperatures and capabilities of the turbo-Brayton cycle change with the demands of the times, and technological development has been continuously pursued to meet such expectations.

Refrigerators used in commercial systems, such as superconductivity power equipment and hydrogen-related equipment, are required to be compact and consume less power, so technology to optimize the refrigeration cycle is important. Also our turbo-Brayton cycle refrigerator (turbo-Brayton refrigerator) has been continuously improved since its launch. As a result, the refrigeration cycle was designed based on the assumption of a single-component refrigerant conventionally, but recently we have found that the use of a multiple-component refrigerant (mixture refrigerant) expands the options for optimization in the design of the refrigeration cycle. This has enabled design that improve refrigerator efficiency while maintaining cooling capacity.

This paper reports on the concept, advantages, and specific consideration results of the utilization of mixture refrigerants in a turbo-Brayton cycle.

### 2. Basic configuration of turbo-Brayton cycle

Figure 1 shows the flow of a turbo-Brayton refrigerator and an ideal T-S diagram of the turbo-Brayton cycle. A turbo-Brayton refrigerator consists mainly of a turbo compressor that adiabatically compresses a refrigerant, an expansion turbine that adiabatically expands the compressed refrigerant to generate cold, a main heat exchanger that exchanges heat between refrigerants to recover cold heat, a sub-cool heat exchanger that exchanges heat between the refrigerant and the fluid being cooled (e.g., liquid nitrogen), and a watercooled cooler that cools the refrigerant by exchanging heat with the cooling water. The equipment and piping that are subject to low temperatures are accommodated in a vacuum insulated tank (cold box). As cooling progresses, the density of the refrigerant decreases and the pressure in the system drops, so a neon gas tank is installed to replenish refrigerant.

The numbers in the T-S diagram in Figure 1 correspond to the flow diagram numbers:  $1\rightarrow 2$  is adiabatic compression,  $3\rightarrow 4$  is isobaric cooling,  $4\rightarrow 5$  is adiabatic expansion, and  $6\rightarrow 1$  is isobaric heating.  $2\rightarrow 3$  is the cooling process by the water cooler and  $5\rightarrow 6$  is the heat absorption process from liquid nitrogen in the sub-cool heat exchanger. In an actual refrigerator, as the adiabatic efficiency of rotating machinery is 70% to 90%, the  $1\rightarrow 2$ and  $4\rightarrow 5$  processes are shown in the diagram with an upward sloping curve and a downward sloping curve, respectively.

Our turbo-Brayton refrigerator is based on the refrigeration cycle shown in Figure 1 and uses a two-stage turbo compressor. This reduces the pressure ratio per stage to achieve high efficiency and also reduces the heat of compression of the refrigerant gas <sup>6</sup>). Currently, we have two models available: a refrigerator with a cooling capacity of 2 kW at a cooling temperature of 69 K (product name: NeoKelvin<sup>®</sup>-Turbo 2kW) and a refrigerator with a cooling capacity of 10 kW at a cooling temperature of 69 K (product name: NeoKelvin<sup>®</sup>-Turbo 10 kW) (Figure 2).



Figure 1 Simple system diagram and ideal T-S diagram of turbo-Brayton cycle



Figure 2 Externals of NeoKelvin<sup>®</sup>-Turbo (a) 2 kW type, (b) 10 kW type

# 3. Conventional refrigeration cycle design method

Our turbo-Brayton refrigerator uses neon as a refrigerant for two reasons. The first is to prevent refrigerant from liquefying in the refrigeration cycle. If refrigerant liquefies at the outlet of the expansion turbine at the cooling temperature of the refrigerator, it causes abnormal vibration of the expansion turbine, which interferes with stable operation. There are three gases that do not liquefy at around 63 K, the cooling temperature: hydrogen, helium, and neon.

The second is the influence of refrigerant physical properties on the design of turbomachinery. For example, in the case of a turbo compressor, the larger the molecular weight of the gas, the higher the pressure ratio  $(P_2/P_1)$  in the T-S diagram in Figure 1) provided that the impeller outer diameter and rotation speed are the same. The NeoKelvin®-Turbo 2kW turbo compressor can only compress to a pressure ratio of about 1.2 with 100% helium at the rotation speed that results in a pressure ratio of 2.0 with 100% neon. To obtain the same pressure ratio of 2.0 as the case of 100% neon using 100% helium, the number of turbo compressors needs to be increased or the impeller outer diameter needs to be increased. However, increasing the number of turbo compressors is undesirable because it causes an increase in the size and cost of the equipment, and increasing the impeller outer diameter leads to an increase in the centrifugal stress of the impeller, causing a problem with the centrifugal strength. Therefore, neon, which has a large molecular weight and enables a higher pressure ratio, was adopted.

It has been reported that the turbo-Brayton cycle is most efficient when the pressure ratio is 2 to 3 <sup>7</sup>). Our turbo-Brayton refrigerator employs a design pressure ratio of 2. The higher the pressure ratio, the higher the expansion ratio of the expansion turbine (ratio of pressure at the expansion turbine outlet to pressure at the expansion turbine inlet), which increases the cold generated and increases the temperature difference between the expansion turbine inlet and outlet ( $T_m$ - $T_c$  in the T-S diagram in Figure 1). Figure 3 shows the relationship between the pressure ratio and the temperature difference between the expansion turbine inlet and outlet. In the figure, we estimated the adiabatic efficiency of the expansion turbine as 80% and that of the turbo compressor as 70% assuming a refrigeration cycle with a liquid nitrogen cooling temperature of 68 K, a liquid nitrogen flow rate of 30 L/min, and a cooling capacity of 2.0 kW. Figure 3 shows that the difference of neon and helium in the temperature difference between the expansion turbine inlet and outlet at a pressure ratio of 2 is less than 4%, indicating that the effect of refrigerant type is minor.

Figure 4 shows the relationship between pressure ratio and expansion turbine outlet temperature under the same estimation conditions as Figure 3. Since the freezing point of liquid nitrogen is approximately 63 K, a pressure ratio of 1.74 or higher may cause the liquid nitrogen to freeze in the sub-cool heat exchanger, resulting in blockage. Therefore, a measure to prevent liquid nitrogen from freezing is necessary for NeoKelvin<sup>®</sup>-Turbo 2kW with the pressure ratio of 2.

With that view, a two-stage sub-cool heat exchanger was adopted with a double-loop structure: parallel-flow heat exchange between neons in the first stage and counter-flow heat exchange between neon and liquid nitrogen in the second stage. Figure 5 shows a schematic flow diagram of a double-loop type sub-cool heat exchanger. The first stage raises the temperature of the neon gas by approximately 4 K, and the temperature of the second-stage heat exchanging area increases above the freezing point of liquid nitrogen <sup>8,9</sup>). However, the double-loop design lengthens the flow path on the neon side, which increases the pressure drop and reduces the cooling capacity by approximately 1%. The pressure ratio was set to 2 to obtain high efficiency of the refrigerator, but the double loop of the sub-cool heat exchanger for preventing the liquid nitrogen from freezing brought about a challenge in improving the performance.



Figure 3 Change of expansion turbine inlet/outlet temperature difference due to pressure ratio (ET: Expansion turbine, Comp.: Compressor)









### 4. Design method using mixture refrigerant

Conventional design first optimizes the refrigeration cycle and then takes measures to prevent freezing according to the temperature, flow rate, heat load, and other conditions of the fluid to be cooled. On the other hand, the newly established refrigeration cycle design method determines the refrigeration cycle so that the entire refrigeration system, including the conditions of the fluid to be cooled, is optimized.

Specifically, first the expansion turbine outlet temperature is set to be above the freezing point of the fluid to be cooled, making the double loop of the sub-cool heat exchanger unnecessary. Next, the inlet temperature at the cold end of the main heat exchanger (point 6 in Figure 1) is set below the inlet temperature of the fluid to be cooled as a condition for the sub-cool heat exchanger to be established. The inlet temperature of the fluid to be cooled is determined from the outlet temperature of the fluid to be cooled, the inlet/outlet pressure, and the cooling capacity. Then, setting the assumed adiabatic efficiency of the expansion turbine determins the expansion turbine inlet temperature. As shown in Figure 3, the pressure ratio of the refrigeration cycle is determined from the expansion turbine temperature difference, and the refrigerant flow rate of the refrigeration cycle is determined from the pressure ratio and cooling capacity. Finally, the refrigerant is selected so that the turbomachinery can obtain high adiabatic efficiency. The following paragraphs describe an example of utilizing a mixture refrigerant of neon and helium for a refrigeration cycle at a cooling temperature of 68 K and a cooling capacity of 2 kW using this design method, and a study of the refrigeration cycle in consideration of the effect on the specific speed of the turbomachinery.

First, to make the double loop of the sub-cool heat exchanger unnecessary, the expansion turbine outlet temperature was set to 63.2 K, which is slightly higher than the freezing point of liquid nitrogen. Assuming a liquid nitrogen flow rate of 30 L/min, the liquid nitrogen inlet temperature is 70.4 K at a cooling capacity of 2 kW. For this reason, the inlet temperature at the cold end of the main heat exchanger was set to 70.1 K, 0.3 K lower than the liquid nitrogen inlet temperature. Assuming the adiabatic efficiency of the expansion turbine to be 80%, the expansion turbine inlet temperature is 73.16 K. Accordingly, the temperature difference at the expansion turbine is 9.96 K, and the pressure ratio is 1.73 as shown in Figure 3. Even if the pressure ratio is reduced, the temperature difference at the expansion turbine is almost the same regardless of the refrigerant type as shown in Figure 3.

Next, Equation (1) shows the formula for the cooling capacity.

$$Q_{ref} = \Delta \mathbf{h} \times \dot{m} \tag{1}$$

where  $Q_{ref}$ : cooling capacity [kW],  $\Delta h$ : enthalpy difference at sub-cool heat exchanger inlet/outlet [kJ/kg], and  $\dot{m}$ : refrigerant mass flow rate [kg/s].

Since the sub-cool heat exchanger outlet temperature (inlet temperature at the cold end of the main heat exchanger) is fixed, lowering the pressure ratio increases the sub-cool heat exchanger inlet temperature (expansion turbine outlet temperature), resulting in a smaller  $\Delta h$ . For this reason, it is necessary to increase the refrigerant mass flow rate according to Equation (1) to compensate for the reduced cooling capacity due to the lower pressure ratio. One of the design values that determine the adiabatic efficiency of turbomachinery is the specific speed. Equation (2) shows the formula for specific speed.

$$N_s = \frac{n\sqrt{Q}}{H^{0.75}} \tag{2}$$

where  $N_s$ : specific speed [rad/s, m<sup>3</sup>/s, J/kg], n: rotational speed [rad/s], Q: refrigerant volume flow rate [m<sup>3</sup>/s], H: theoretical adiabatic head [J/kg].

The units used to express specific speed are generally different between turbo compressors and expansion turbines, and the ones for expansion turbines are used here. The units used in turbo compressors are Ns: specific speed [rpm, m<sup>3</sup>/min, m], n: rotational speed [rpm], Q: refrigerant volume flow rate [m<sup>3</sup>/min], and H: theoretical adiabatic head [m]. The specific speeds with the highest adiabatic efficiency are said to be 0.6 to 0.7 [rad/s, m<sup>3</sup>/s, J/kg] for expansion turbines and 300 to 400 [rpm, m<sup>3</sup>/min, m] for turbo compressors <sup>10</sup>.

Here, when the refrigerant is changed while keeping the rotational speed n constant, the refrigerant volume flow rate Q and theoretical adiabatic head H increase or decrease due to differences in physical properties, resulting in a change in the specific speed. With a single-component refrigerant, the specific speed is uniquely determined from the operating conditions, but with a mixture refrigerant, the specific speed can be changed without changing the rotational speed n by adjusting the concentration.

Figure 6 shows the temperature difference between the expansion turbine inlet and outlet when the adiabatic efficiency of the turbo compressor and the expansion turbine are increased by 3% each. As the adiabatic efficiency increases, the temperature at the outlet of the expansion turbine decreases at the same pressure ratio, so it is necessary to check again that the pressure ratio does not cause liquid nitrogen to freeze. As a result, a pressure ratio of 1.7 was adopted in this study.



Figure 6 Comparison of expansion turbine outlet temperature due to adiabatic efficiency (ET: Expansion turbine, Comp.: Compressor)

Figure 7 shows the relationship between helium concentration and expansion turbine specific speed at pressure ratios of 2.0 and 1.7. In the case of using 100% neon refrigerant, the mass flow rate with which the cooling capacity of 2.0 kW is obtained is 0.2 kg/s at a pressure ratio of 2.0 and 0.275 kg/s at a pressure ratio of 1.7. When the helium concentration of the refrigerant is increased at a pressure ratio of 1.7, the mass flow rate with which a cooling capacity of 2.0 kW is obtained is 0.054 kg/s.

Figure 7 shows that the refrigeration cycle with a pressure ratio of 1.7 falls within the optimum specific speed range of 0.6 to 0.7 when the helium concentration is between 0 and 23%. In particular, when a specific speed of 0.65, which is at the center of the optimal range, is aimed for, a helium concentration of 11% is optimal. A helium concentration of 11% will improve the specific speed of the turbo compressor and expansion turbine, which can be expected to improve the adiabatic efficiency by about 3% each <sup>10</sup>).



Figure 7 Change in specific speed due to helium concentration (Nm<sup>3</sup>/h : m<sup>3</sup>/h at 273.15 K, 101.35 kPa, ET: Expansion turbine, Comp.: Compressor)

On the other hand, the turbo compressor power also increases because the volume flow rate increases compared to the conventional refrigeration cycle, and if the efficiency of rotating machinery is not improved, the coefficient of performance (COP) of the refrigerator will decrease. Equation (3) shows the formula for calculating COP.

$$COP = \frac{Q_{ref}}{W}$$
(3)

where W: power consumption [kW] of the turbo compressor.

Table 1 compares the refrigeration cycle specifications between neon and mixture refrigerant. Based on the comparison results, we were able to establish a refrigeration cycle that does not require a double loop of the sub-cool heat exchanger and improves the COP by approximately 1%.

Furthermore, mixing helium also improves the heat transfer performance at the heat exchanger and reduces the pressure loss, which are factors not included in Table 1. By redesigning the heat exchanger and piping according to the pressure and flow rate of the mixture refrigerant cycle, the cooling capacity and COP can be improved beyond the results shown in Table 1.

Figure 8 shows the ideal T-S diagram in which blue lines are used to indicate the refrigeration cycle that uses the low pressure ratio and the mixture refrigerant. The numbers in the diagrams that are changed from the conventional refrigeration cycle (red line) are marked with a dash for comparison.

Table 1 Comparison of specifications between Neon cycle and mixture refrigerant cycle (ET : Expantion turbine, Comp. : Compressor)

	Unit	Neon cycle	Mixure refrigerant cycle
Helium concentration	[mol%]	0	11
Pressure ratio	[-]	2.0	1.7
Refrigerant flow rate	[Nm <sup>3</sup> /h]	800	1100
ET outlet temperature	[K]	60.9	63.2
ET specific speed	[-]	0.47	0.65
Comp. specific speed	[-]	112	151
ET adiabatic efficiency	[%]	80	83
Comp. adiabatic efficiency	[%]	70	73
Cooling capacity	[kW]	2.0	2.0
Comp. consumption	[kW]	26.9	26.5
COP	[-]	0.0757	0.0764



Figure 8 Ideal T-S diagram of turbo-Brayton cycle under the low pressure ratio and mixture refrigerant condition

### 5. Conclusion

Through the practical application of the turbo-Brayton cycle and subsequent continuous efforts to improve performance, we established a method for design optimization of a refrigeration cycle using mixture refrigerant and turbomachinery. Using this method, we have shown a design method of a refrigeration cycle that can prevent the fluid to be cooled from freezing in the sub-cool heat exchanger while improving the COP of the refrigerator.

As an example, a refrigeration cycle using neon refrigerant and mixture refrigerant was studied with a refrigerator having a cooling capacity of 2 kW. As a result, a refrigeration cycle with a pressure ratio of 1.7 eliminated the need for a double loop of sub-cool heat exchanger. Furthermore, the expansion turbine specific speed was optimized by setting the helium concentration to 11%, which improved the adiabatic efficiency of the expansion turbine and increased the COP by approximately 1% compared to the refrigeration cycle using neon.

The method of designing a refrigerator with a mixture refrigerant reported here was patented in 2021 <sup>12</sup>). The mixture refrigerant in this design method is not limited to a combination of neon and helium, but any gas can be selected according to the cooling temperature. For example, for reliquefying LNG, since its cooling temperature is approximately 110 K, nitrogen and argon can be used for the mixture refrigerant. In addition, neon liquefies at 30 K, but using a mixture refrigerant can extend the cooling temperature range to 25 K or lower without causing liquefying.

Technologies for refrigeration cycles and turbomachinery have developed in response to the demands of the times. Based on the research results of this time on the turboBrayton cycle using a mixture refrigerant, we will actively search for new applications and develop and commercialize new technologies so as to contribute to solving recent social issues such as energy conversion and carbon neutrality.

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