Thermodynamic comparison of small liquid nitrogen generators driven by mixed-refrigerant J-T refrigerators and gas expansion cycles

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Abstract. The thermodynamic comparison of small mixed-refrigerant J-T (MRJT) and gas expansion (Kapitza cycle) LN2 generators are conducted by the exergy method in this paper. Both of two types are employed for low pressure N2 liquefaction and air separation (<9.0 bar). Pure N2, PSA units or cryogenic air separation columns are used for LN2 production. The MRJT type is driven by single-stage compressors. It is indicated that the efficiency of MRJT type is higher than Kapitza type. With pure N2 at 8.0 bar, the figures of merit (FOM) of MRJT and Kapitza types are 26.86% and 12.79%, respectively. With mini air separation columns, the FOM of MRJT and Kapitza types are 21.42% and 11.34%, respectively. The large exergy losses in the compressor unit and non-isentropic expansion are the main reasons of the inferior efficiency of the Kapitza type. In addition, the Kapitza type requires larger total compressor displacement than the MRJT type. The MRJT type is recommended to be used in small LN2 generators.

1. Introduction

Liquid nitrogen (LN2) is a widely used cryogenic fluid. Small LN2 generators are convenient choices for field and remote users, or laboratories with small-quantity-high-frequency LN2 requirements. The cooling power for N2 liquefaction could be supplied by helium regenerative cryocoolers (Stirling/G-M, etc.), mixed-refrigerant J-T (MRJT) refrigerators or feed gas expansion cycles (Kapitza cycle [1], etc.). Although Stirling/G-M cryocoolers achieve relatively high efficiencies with compact configurations [2-4], their high-grade cooling power is only utilized at the cold head to cool down the feed gas from ambient temperature. Besides, their cost might be higher than MRJT or Kapitza systems [5].

As a contrast, the feed gas is cooled down along the recuperator at various temperatures in MRJT and Kapitza types. MRJT refrigerators could cover cooling requirements at 80-200 K with high efficiency and low hardware cost, especially for cooling temperature-distributed heat loads like gas cooling and liquefaction [6-7]. Several MRJT LN2 liquefiers have been investigated with high (≥20 bar) [8-10] and low pressure (8 bar) feed N2 [11-14], similar with MRC natural gas liquefaction processes. Feed gas recuperative expansion processes are main cooling methods for large scale air liquefaction/separation plants. Based on Kapitza cycles for low pressure gas liquefaction, small LN2 generators [15-16] are developed with simple configurations. However, detailed comparisons of Kapitza and MRJT types are rarely reported. Therefore, the thermodynamic performance of these two types of LN2 generators are compared in this paper, which could be a reference for the design of small liquefiers.

2. Process configuration

The LN2 generators in this paper are cooled by MRJT refrigerators and Kapitza cycles respectively. Both of these two types are employed for low pressure pure N2 liquefaction and air separation.
2.1. MRJT type LN$_2$ generator

The MRJT LN$_2$ generators are based on single-stage MRJT separation cycles, which are precooled by normal propane vapor-compression cycles, as shown in figure 1 and 2. With bottled pure N$_2$ or a PSA (pressure swing adsorption) unit as the gas source, compressed N$_2$ is directly liquefied by MRJT cycle, as Process A in figure 1. After being precooled by propane in precooling heat exchanger (HX1), feed N$_2$ and high-pressure warm Ne-N$_2$-HC mixed-refrigerant is cooled by low-pressure cold mixed-refrigerant in recuperators (HX2 and HX3). LN$_2$ is throttled to 1.0 bar as product. The boiled-off gas (BOG) is not recovered to simplify the configuration of the small system. Both the main and precooling cycles are driven by single-stage oil-lubricated compressors for normal refrigeration, with discharge pressures below 20 bar (all pressures are absolute value) and pressure ratio less than 6. Liquefaction pressures of N$_2$ are within the permissible range of normal small air compressors and PSA units (< 9.0 bar).

Figure 1. Process configuration of MRJT LN$_2$ generator with pure feed N$_2$ (Process A)

Beside using pure N$_2$ or a PSA unit, LN$_2$ could also be produced by a mini air separation rectification column. Feed air is compressed, partly liquefied by the refrigerator and separated in the column, as shown in Process C of figure 2. In order to avoid the safety problems in the intermediate-wall heat transfer between the flammable Ne-N$_2$-HC mixed-refrigerant and air (21% of O$_2$), a closed N$_2$ cycle is set to convey the cooling capacity from the MRJT refrigerator to the air. The circulating N$_2$ is liquefied by the same MRJT refrigerator as Process A. Compressed feed air is cooled to two-phase status by the throttled circulating N$_2$ in HX4 and separated by a packed column. LN$_2$ is gained at the column top. The column is only designed for stable LN$_2$ production, thus the reboiler and stripping section are removed to simplify its configuration [12]. The cooling power for the condensation of the nitrogen-enriched vapor in the top condenser is provided by the throttled bottom liquid (oxygen-enriched air, throttled to 1.7 bar). The rest of the cooling capacity of the oxygen-enriched air is recovered in HX4 and released as waste.

Figure 2. Process configuration of MRJT LN$_2$ generator with air separation column (Process C)
2.2. Kapitza LN2 generator
Kapitza cycle are typical low-pressure gas expansion liquefaction process, which are applied in small liquefaction systems. For pure N2 liquefaction, compressed feed N2 is cooled and liquefied by the recuperation, expansion and throttling processes. Most of the N2 is cycling in the loop and performing as refrigerant, as Process E in figure 3. A nonflammable R22 precooling cycle is employed to precool N2 to 255 K before the main recuperator (HX3). The high-pressure warm N2 is cooled down by low pressure cold N2 in HX3 and liquefied in HX4. The liquid is throttled and separated in the cold flash separator. Most of the feed gas is sent to the expander before HX4 and mixed with flashed vapor to provide the cooling capacity for liquefaction.

![Figure 3. Process configuration of Kapitza type LN2 generator with pure feed N2 (Process E)](image)

For air separation, air is also cooled by a Kapitza cycle, as Process F in figure 4. Compressed feed air is precooled in HX2 and cooled to its two-phase state in HX3 and HX4 before entering the column. The column in Process F is the same as that in Process C, which is also only designed for LN2 production. The throttled oxygen-enriched air and expanded air are mixed and sent into recuperators (HX4, HX3 and HX1) to provide the cooling capacity, then released as waste gas.

![Figure 4. Process configuration of Kapitza type LN2 generator with air separation column (Process F)](image)

3. Thermodynamic analysis methods

3.1. Analysis methods
LN2 output \( (g_{v, LN2}) \), specific power consumption \( (SPC) \) and figure of merit \( (FOM) \) are the main evaluation parameters. The \( SPC \) is the ratio of the LN2 volume flow rate to the overall power consumption \( (W_{total}) \), as equation (1); The \( FOM \) is the ratio of exergy gained by the feed gas to \( W_{total} \), also the exergy efficiency of system, as equation (2). The exergy gained by feed gas is the exergy difference between the inlet feed N2/air and outlet LN2. The expansion power \( (W_{EP}) \) is completely recovered. Detailed methods are listed in [12].

\[
SPC = \frac{g_{v, LN2}}{W_{total}} = \frac{g_{v, LN2}}{W_{CP, main} + W_{CP, precooling} - W_{EP}} \\
FOM = \frac{Minimum \ liquefaction \ power}{W_{total}} = \frac{m_{LN2}(e_{LN2, out} - e_{feed, in})}{W_{CP, main} + W_{CP, precooling} - W_{EP}}
\]
Exergy losses and exergy loss fractions are calculated by exergy balance equations, as equation (3-4).

\[ I_j = \sum E_{in} - \sum E_{out} + \sum W_{input} - \sum W_{output} - \int_0^\infty (T_0/T - 1)\delta Q_c \]  
\[ \Pi_j = \frac{I_j}{W_{total}} \sum \Pi_j + FOM = 1 \]

3.2. Simulation conditions
The simulation conditions are set according to the operating range of normal refrigeration hardware. The minimum approaches in heat exchangers (\(\Delta T_{min}\)) are 3 K. The maximum compressor discharge temperature is 385.15 K; liquid in the compressor is not allowed. The aftercooler outlet temperature and feed gas temperature are 308.15 K. The maximum discharge and minimum suction pressures of compressors are 20 bar and 2 bar, respectively. The pressure drops of both high- and low-pressure streams are less than 1.0 bar. The adiabatic efficiencies of the compressors and expanders are 60% and 70% respectively. The ambient temperature is 300 K. The mixture composition shift and heat leak are ignored. The mixture properties are calculated by the PR-vdW model [17-18]. The \(FOM\) is the only optimization target in this simulation, in keeping with the research reported in [12].

4. Performance analysis and comparison

4.1. Performance of MRJT type LN\(_2\) generators
The key operating parameters of Process A (MRJT + Pure N\(_2\)) and C (MRJT + Column) are listed in table 1. All operating pressures of the main, precooling and cycling N\(_2\) compressors are within the range of single-stage compressors for normal refrigeration. The cold end temperature of the MRJT refrigerator is 98 K. The main mixed-refrigerant consists of Ne, N\(_2\), CH\(_4\), CF\(_4\), C\(_2\)H\(_6\), C\(_3\)H\(_8\) and iC\(_4\)H\(_{10}\).

| Table 1. Operating parameters in MRJT type LN\(_2\) generators |
|---------------------------------------------------------------|
| Process | MRJT + Pure N\(_2\) (A) | MRJT + Column (C) |
| --- | --- | --- | --- |
| Cycle | Main | Precooling | Main | Precooling | N\(_2\) |
| Compressor suction pressure (bar) | 3.50 | 2.20 | 3.50 | 2.20 | 4.70 |
| Compressor discharge pressure (bar) | 17.00 | 12.50 | 17.00 | 12.50 | 8.20 |
| Temperature before throttling (K) | 98.00 | 308.15 | 98.00 | 308.15 | 98.00 |
| Temperature after throttling (K) | 90.43 | 252.67 | 90.43 | 252.67 | 94.29 |

The molar compositions of refrigerants in MRJT LN2 generator are listed in table 2. The composition along the whole cycle is assumed to be unchanged.

| Table 2. The molar compositions of refrigerants in MRJT type LN\(_2\) generators |
|---------------------------------------------------------------|
| Component (%) | Ne | N\(_2\) | CH\(_4\) | CF\(_4\) | C\(_2\)H\(_6\) | C\(_3\)H\(_8\) | iC\(_4\)H\(_{10}\) |
| Main cycle | 3.9 | 32.1 | 24.6 | 8.1 | 11.9 | 12.8 | 6.6 |
| Precooling cycle | / | / | / | / | / | 100 | / |

As shown in the T-Q diagrams of figure 5, the match of cold and warm streams in the recuperator of the MRJT refrigerator (HX1-HX3) is relatively favorable due to the optimization of mixture composition. A small recuperation exergy loss could be achieved with reduced temperature difference. The optimized operating pressure of N\(_2\) cycle could also reduce the temperature differences in the air liquefaction heat exchanger (HX4), which could decrease the exergy losses in it.
The mini column could achieve superior performance at lower separation pressures ($p_{\text{air}, \text{sep}}$). The minimum $p_{\text{air}, \text{sep}}$ is 3.87 bar to ensure the nitrogen-enriched vapor at the column top can be condensed by the throttled bottom liquid. However, the efficiency of the air liquefaction unit is higher under higher liquefaction pressures ($p_{\text{air}, \text{h}}$), as analyzed in [12]. Thus, the two-phase air is throttled before entering the column, achieving high-pressure liquefaction and low-pressure separation. The minimum SPC of the MRJT + Column type is 0.78 kWh/L with $p_{\text{air}, \text{sep}} = 3.9$ bar and $p_{\text{air}, \text{h}} = 6.4$ bar, as shown in figure 6.

**Figure 5.** $T$-$Q$ diagrams of each heat exchanger in Process A and C

4.2. Performance of Kapitza type LN$_2$ generators

The key operating parameters of the Kapitza + Pure N$_2$ (Process E) and the Kapitza + Column (Process F) are listed in table 3. The feed gas is compressed from 1 bar to 8 bar by normal small air compressors. Most of the feed gas (> 80%) is sent to the expander to generate the cooling capacity for liquefaction.

**Table 3.** Operating parameters in gas expansion (Kapitza type) LN$_2$ generators

| Process          | Kapitza + Pure N$_2$ (E) | Kapitza + Column (F) |
|------------------|--------------------------|----------------------|
| Cycle            | N$_2$                    | Precooling           | Air Precooling |
| Compressor suction pressure (bar) | 1.00 | 2.27 | 1.00 | 2.27 |
| Compressor discharge pressure (bar) | 8.00 | 14.00 | 8.00 | 14.00 |
| Temperature before expansion (K)   | 116.05 | 308.15 | 121.79 | 308.15 |
| Temperature after expansion (K)    | 83.46 | 251.96 | 87.73 | 251.96 |
Compared with the MRJT types, the temperature difference ($\Delta T$) in the recuperator of the Kapitza type is relatively large (especially at low temperatures), leading to a large recuperation exergy loss. Since the minimum approach ($\Delta T_{\text{min}}$) appears at the warm end of each recuperator, the R22 precooling cycle could push $\Delta T_{\text{min}}$ to lower temperatures, avoiding $\Delta T$ being enlarged further and improving efficiency.

**Figure 7.** $T$-$Q$ diagrams of each heat exchanger in Process E and F

For pure N$_2$ liquefaction, a larger LN$_2$ output and higher FOM could be achieved, as illustrated in figure 8. However, the discharge pressure of the small air compressors and PSA unit is limited. For air separation, a low SPC could be achieved under high liquefaction pressures (at high $p_{\text{air,h}}$) but low separation pressures ($p_{\text{air,sep}}$), with a lower liquefaction power consumption and higher product ratio. The maximum FOM is 11.34% with $p_{\text{air,sep}}$ = 3.9 bar and $p_{\text{air,h}}$ = 8.0 bar, and SPC = 1.48 kWh/L.

**Figure 8.** FOM and LN$_2$ yield of Process E under various N$_2$ compressor discharge pressures

**Figure 9.** The FOM and product ratio of Process F under various $p_{\text{air,h}}$ and $p_{\text{air,sep}}$

### 4.3. Performance comparison of MRJT and Kapitza types of LN$_2$ generators

The exergy loss fractions of each component in the MRJT and Kapitza LN$_2$ generators are illustrated in figure 10. For the MRJT type, besides the compressors, the recuperators occupy the largest exergy loss. For the Kapitza type, the exergy losses in the after cooler are relatively large due to the high adiabatic exponent of N$_2$ and air, which leads to high discharge temperatures. Additionally, the exergy losses in the expanders of the Kapitza type are obviously larger than those in the J-T element of the MRJT type. In actual mini Kapitza systems, the wasted expansion work would enlarge the expansion exergy loss further.
The overall comparison is shown in figure 11. Whether for pure N\textsubscript{2} liquefaction or air separation, the SPC and FOM of MRJT types are obviously superior to Kapitza types. Thus, the operating cost of MRJT types might be lower than Kapitza types. Besides, for a certain LN\textsubscript{2} output, a smaller total compressor motor power and displacement are required in MRJT types, reducing the construction cost of the compressor. Although the total heat exchanger UA value of the Kapitza type is smaller, the heat transfer coefficient (U) of a mixed-refrigerant might be higher than N\textsubscript{2} or air. Thus, the actual heat transfer area (A) of the MRJT type might be similar to that of the Kapitza type, as well as the construction cost of the heat exchanger. Besides, the cost of a J-T element might be lower than the cryogenic high-speed component of an expander. Therefore, the total construction of MRJT types could be lower than Kapitza types.

![Figure 10. Exergy loss distributions (II) in Process A/C/E/F](image)

The experimental SPC of the MRJT type was 2.01 kWh/L with pure N\textsubscript{2} at 8.0 bar (1.12L/h, compression power considered) and 4.59 kWh/L with the air separation column (0.60 L/h, purity of 97.5 %), as reported in [13]. In contrast, the SPC of the Kapitza type pure N\textsubscript{2} liquefier in [16] was 2.12 kWh/L (17.4 L/h); the small air separation system in [15] was 4.28 kWh/L (28.0 L/h). The MRJT types could achieve lower or similar SPC with Kapitza types under much smaller LN\textsubscript{2} producing capacity and system scale. In addition, small scale in this paper is corresponding to the LN\textsubscript{2} generators with LN\textsubscript{2} output of 0.1 L/h-200 L/h approximately, mainly 1 L/h-10 L/h, which could be movable and used in labs.

![Figure 11. Exergy loss distributions (II) in Process A/C/E/F](image)
5. Summary and discussion
The thermodynamic comparison of small MRJT type and gas expansion (Kapitza type) LN$_2$ generators are conducted in this paper. Both for low-pressure (< 9.0 bar) N$_2$ liquefaction and air separation, it is indicated that the efficiencies of MRJT types are higher than Kapitza types. With pure N$_2$ at 8.0 bar, the FOM of MRJT and Kapitza types are 26.86% and 12.79%, respectively. With air separation columns, the FOM of MRJT and Kapitza types are 21.42% and 11.34%, respectively. The large exergy losses in the non-isentropic expansion and after cooler are the main reasons of the inferior efficiency of the Kapitza type. Besides, the Kapitza type requires a larger total compressor displacement and construction cost than the MRJT type. Therefore, the MRJT type is more favorable for small LN$_2$ generators.

6. References
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