Development of cryogenic cooling system using a double G-M cryo-cooler for NMR spectrometer

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Abstract. The sensitivity of nuclear magnetic resonance (NMR) spectrometer is limited by thermal noise. To increase the signal-noise-ratio (SNR), cryogenically cooled probes with cryo-coolers have been used. In order to cool RF coils in the probe below 10K, Gifford-McMahon/Joule-Thomson (G-M/J-T) cryo-coolers are used. But, the G-M/J-T cryo-coolers aren’t suitable for a long time operation. On the other hand, G-M cryo-coolers have high reliability. We designed a cryogenically cooled probe system using a double G-M cryo-cooler with an RF coil and preamplifiers that could be cooled below 5 and 50K, respectively. This probe system consists of a probe unit, a transfer unit and a cryo-cooling unit. The cryo-cooling unit consists of two G-M cryo-coolers, two counter-flow heat exchangers, helium gas lines, etc. Compressed helium gas is cooled by the cooling stages of G-M cryo-coolers and two heat exchangers. The cooled helium gas is supplied to the probe unit through the 2.5m long transfer unit which consists of a 45mm diameter flexible vacuum vessel containing four helium gas lines. Cold stages for the RF coil and the preamplifiers in the probe unit are cooled by the cryogenically cooled helium gas. Experimental results showed that the cold stage temperature for the RF coil and for preamplifiers after 12 hours was 5.29 and 40.3K, respectively.

1. Introduction
Nuclear magnetic resonance (NMR) spectrometer is a well-known technique for the static analysis of molecules in an equilibrium state. It is used to analyze the molecular structures of amino acids and proteins. In the field of NMR spectrometer, a new NMR spectrometer with the improved time-resolution suitable for dynamic analysis is necessary. To perform dynamic NMR analysis, high sensitivity is needed so that NMR spectra can be rapidly measured. The sensitivity of an NMR spectrometer is limited by thermal noise originating from RF coils and preamplifiers in the probe. Using a cryogenically cooled probe, it can improve the signal-noise-ratio (SNR) in the NMR spectrometer. The SNR using the cryogenically cooled probe is 4 times of that using a probe on room temperature in case an RF coil is cooled to temperature between 10 and 77K [1].

At first, liquid helium was used as a cryogenic cooling source [2, 3, 4]. However, the running costs were too expensive to perform NMR spectrometer for a long period of time. On the other hand, a
A closed-cycle cooling system using cryo-cooler was developed for cooling cavity maser below 4 K [5, 6, 7]. This system consisted of a two-stage Gifford-McMahon (G-M) cryo-cooler and heat exchangers. Joule-Thomson (J-T) valves were used to improve the cooling power compared with the original cooling capacity of the cryo-cooler. Yokota et al. demonstrated a 4.5K-cooling system for a cryogenically cooled probe that used a G-M/J-T cryo-cooler [8, 9]. They described that if RF coils are cooled to 4.5K, the gain in SNR is double that of a 20K-cryogenic probe.

In order to regulate the cooling power using a G-M/J-T cryo-cooler, delicate operations of J-T valve are necessary. A J-T valve is a similar to a very fine needle valve and can easily be damaged by overtightening. In closed-cycle operation, contaminants gradually accumulate by freezing out at the J-T valve; therefore, a G-M/J-T cryo-cooler has problems when it is in operation for a long period of time. On the other hand, the G-M cryo-cooler has high-reliability in the field of superconducting magnets or cryopumps. Although, the cooling power of G-M cryo-cooler is lower than that of G-M/J-T cryo-cooler. The purpose of this work is to develop a cooling system for cooling the cold stage with RF coil at 5K, and to improve the reliability when in operation for a long period of time.

**Figure 1.** Schematics of cryogenically cooled probe system

### 2. Structure

We developed an original cryogenically cooled probe system. Figure 1 shows a schematic diagram of a cryogenically cooled probe system. The system consists of a probe unit, a transfer unit, and a cryo-cooling unit. These units were constructed in our workshop. In the probe unit, an RF coil is thermally anchored to a cold stage that is cooled by compressed helium gas. Some preamplifiers are cooled at the same time as the RF coil, but to different temperatures on a different cold stage. We designed a cryogenically cooled probe system using a double G-M cryo-cooler with an RF coil and preamplifiers that could be cooled to 5K and below 50K, respectively.

We measured the temperature at 15 points. Temperatures were measured using calibrated Cernox™ resistance temperature sensors. They are attached to the surfaces of pipes and the cold stage. The mass flow rate of compressed helium gas was set to 0.3 g/s at 1.0 MPa using a mass flow controller.
2.1. Cryo-Cooling Unit

The cryo-cooling unit consists of two G-M cryo-coolers, four heat exchangers on the cold stages of the G-M cryo-coolers, and two counter-flow heat exchangers. We used two commercial G-M cryo-coolers produced by Sumitomo Heavy Industries Ltd. Table 1 shows the specifications of the G-M cryo-cooler. The model used was an RDK-415D, which provided 1.5 W at 4.2K. The 1st stage of the G-M cryo-coolers cools a thermal shield. In this cooling unit, two G-M cryo-coolers are mounted on a vacuum chamber.

We developed and constructed counter-flow heat exchangers in our workshop. Table 2 shows the specifications of these heat exchangers. The 1st heat exchanger transfers heat between areas at 300 and 45K and the 2nd heat exchanger transfers heat between areas at 30 and 6K. The design efficiencies were over 93% for the 1st heat exchanger and over 95% for the 2nd heat exchanger.

There are no J-T valves in this unit. The cooling capacity is equal to the cooling performance of the G-M cryo-coolers.

| Table 1. Specifications of G-M cryo-cooler |
|------------------------------------------|
| Manufacture                             | Sumitomo Heavy Industries Ltd. |
| Model                                    | RDK-415D                       |
| Type                                     | Gifford-McMahon                |
| Cold Stage                               | 2 Stages                       |
| Compressor                               | 7 kW, air cooled               |
| Heat Capacity                            | 35 W at 50K(1st stage)         |
|                                          | 1.5 W at 4.2K (2nd stage)      |

| Table 2. Specifications of counter-flow heat exchangers. |
|---------------|---------------|
| 1st heat exchanger | 2nd heat exchanger |
| Temperature (high) | 300K            | 30K            |
| Temperature (low)  | 45K             | 6K             |
| Length            | 265 mm          | 310 mm         |
| Diameter          | 38 mm           | 42 mm          |

The circuit process is as follows:

1. Compressed helium gas at 1.0 MPa is supplied from the helium compressor. The compressed helium gas at room temperature is cooled to 45K by passing it through the 1st counter-flow heat exchanger.

2. The compressed helium gas is cooled to 30K by passing it through the 1st cold stage of a G-M cryo-cooler.

3. The compressed helium gas is supplied to the probe unit through the transfer unit (50 K helium gas line, Supply). It cools some preamplifiers and thermal shields in the probe unit to about 40K.

4. The warmed helium gas is returned to the cryo-cooling unit through the transfer unit (50K helium gas line, Return) and is cooled to 30K by passing it through another 1st cold stage of a G-M cryo-cooler.

5. The compressed helium gas is cooled to 6K by passing it through the 2nd counter-flow heat exchanger.

6. The compressed helium gas is cooled to 6K by passing it through the 2nd counter-flow heat exchanger.

7. The compressed helium gas is supplied to the probe unit through the transfer unit (5K helium gas line, Supply). In the probe unit the gas cools an RF coil.

8. The warmed helium gas is returned to the cryo-cooling unit through the transfer unit (5K helium gas line, Return). It is used as the low temperature source of the two counter-flow heat exchangers.

9. The helium gas, now warmed to room temperature is returned to the helium compressor.
2.2. Transfer Unit

The cryo-cooling unit must be installed 2.5m from the NMR magnet in order to avoid the magnetic field. Therefore, the transfer unit is to connect the probe unit with the cryo-cooling unit. Figure 2 shows the structure of the transfer unit. The outside surface of the unit comprises a flexible stainless steel pipe that acts as a vacuum chamber. Outside diameter of the flexible stainless steel pipe is 45 mm. There are two copper pipes in the centre of the unit through which helium at 5K is supplied (Supply, Return) to cool the cold stage on which the RF coil is located in the probe unit. There is a flexible helium gas chamber between the vacuum chamber and the 5K helium gas lines. It was charged with helium at 0.1 MPa. Two stainless pipes are rolled spirally inside the flexible helium gas chamber. Inside these stainless pipes, compressed helium gas that is cooled below 50K flows. These stainless steel pipes are gas lines (Supply, Return) that supply helium gas at 50K to cool the preamplifiers and thermal shields in the probe unit. Helium gas inside the helium gas chamber is cooled by these helium gas lines acting as a thermal shield.

A super insulator is wound between the flexible helium gas chamber and the flexible vacuum chamber to reduce the radiation heat load.

Figure 2. Schematic of transfer unit.

Figure 3. Temperature history of cryogenically cooled probe system
3. Results and Discussion
Figure 3 shows the temperature history of the cryogenically cooled probe system. When the G-M cryo-coolers were started, the temperatures at some measured points in the cryogenically cooled probe system fell slowly. After 12 hours, the temperature of the cold stage for RF coil and preamplifiers were 5.29 and 40.3K, respectively. We did not have to adjust valves, mass flow rates and the pressure of the circulating helium gas during this 12-hour period.
Table 3 shows the temperature in the cryogenically cooled probe system at a steady state. The pressure of compressed helium gas is at 0.65 MPa because the density of helium gas decreases at low temperatures. The heat input between the temperature measuring points was calculated as follow:

\[ Q = m \cdot (h_2 - h_1) \]

where \( Q \) is heat load [W], \( m \) is mass flow rate [kg/s] of circulated helium gas, and \( h \) is enthalpy [J/kg] of helium gas calculated using the temperature and pressure at a given measured point. The calculation results are shown in Table 4. The heat load of the 50K helium gas lines (Supply, Return) are 5.1 and 0.5 W, respectively.

| Table 3. Temperature at measuring points |
|-----------------------------------------|
| Measured position | Temperature [K] |
|-------------------|-----------------|
| 1                 | 301.2           |
| 2                 | 40.9            |
| 3                 | 32.4            |
| 4                 | 35.6            |
| 5                 | 40.3            |
| 6                 | 40.6            |
| 7                 | 31.2            |
| 8                 | 6.27            |
| 9                 | 4.98            |
| 10                | 3.80            |
| 11                | 4.08            |
| 12                | 5.29            |
| 13                | 5.43            |
| 14                | 30.2            |
| 15                | 286.2           |

| Table 4. Heat load calculation |
|--------------------------------|
| Process | Heat load [W] |
|---------|---------------|
| 1st Heat Exchanger (Supply)  | -397.7        |
| 1st stage of G-M Cryo-Cooler (1) | -18.6         |
| Transfer unit (50K Supply Line) | 5.1           |
| Cold stage for preamplifier | 7.4            |
| Transfer unit (50K Return Line) | 0.5           |
| 1st stage of G-M Cryo-Cooler (2) | -14.8         |
| 2nd Heat Exchanger (Supply)  | -45.9          |
| 2nd stage of G-M Cryo-Cooler (1) | -2.20         |
| 2nd stage of G-M Cryo-Cooler (2) | -1.26         |
| Transfer unit (5K Supply Line) | 0.22           |
| Cold stage for RF coil | 1.46           |
| Transfer unit (5K Return Line) | 0.21           |
| 2nd Heat Exchanger (Return) | 45.8           |
| 1st Heat Exchanger (Return)  | 396.6          |
The heat load of the Supply line was larger than the Return line. In the transfer unit, the 50K helium gas lines are installed in the helium gas charged chamber. In the helium gas charged chamber, heat was exchanged between the 50K helium gas lines (Supply, Return) and the charged helium gas. Therefore, the helium gas supply line received the heat load because the helium gas supply line was at a lower temperature in the helium gas charged chamber. The heat loads of the 5K helium gas lines (Supply, Return) were 0.22 and 0.21W, respectively. That is the reason the heat loads on the 5K helium gas lines (Supply, Return) were essentially equal to the heat transfer between the supply and return lines. The heat load of the cold stage for the RF coil in the probe unit was 1.46 W at 5.29K.

4. Conclusions

We developed a cryogenically cooled probe system for use in an NMR spectrometer. We designed a cryogenically cooled probe system using a double G-M cryo-cooler with an RF coil and preamplifiers that could be cooled below 5 and 50K, respectively. The cryogenically cooled probe system consisted of a cryo-cooling unit, a transfer unit, and a probe unit. The cryo-cooling unit has a double G-M cryo-cooler, four heat exchangers on the cold stages of the G-M cryo-coolers, and two counter-flow heat exchangers. The transfer unit has two pipes that contain 5K helium gas lines (Supply, Return) and two pipes that contain 50K helium gas lines (Supply, Return). The probe unit has cold stages for an RF coil and for preamplifiers. We measured the cooling performance of the cryogenically cooled probe system. The performance of system can be summarised as follows:

1. The cold stage temperature for the RF coil and for preamplifiers after 12 hours was 5.29 and 40.3K, respectively.
2. The present cryo-cooling unit has a cooling capacity of 1.46 W at 5.29K at the cold stage for the RF coil.

Acknowledgments

The authors would like to thank Professors E. Masada, S. Kimura, T. Kohzuma, D. Kohda, and H. Morita for discussions. This work has been supported in part by the Research Promotion Bureau, Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan, under the following contracts: Nos.17–260 and 18–489.

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