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Magnetic refrigerator for hydrogen liquefaction

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Abstract. Magnetic refrigeration which is based on the magnetocaloric effect of solids has the potential to achieve high thermal efficiency for hydrogen liquefaction. We have been developing a magnetic refrigerator for hydrogen liquefaction which cools down hydrogen gas from liquid natural gas temperature and liquefies at 20 K. The magnetic liquefaction system consists of two magnetic refrigerators: Carnot magnetic refrigerator (CMR) and active magnetic regenerator (AMR) device. CMR with Carnot cycle succeeded in liquefying hydrogen at 20K. Above liquefaction temperature, a regenerative refrigeration cycle should be necessary to precool hydrogen gas, because adiabatic temperature change of magnetic material is reduced due to a large lattice specific heat of magnetic materials. We have tested an AMR device as the precooling stage. It was confirmed for the first time that AMR cycle worked around 20 K.

1. Introduction

Recently hydrogen energy is considered as an alternative for fossil fuels because production of greenhouse gases can be reduced. For efficient, economic transport and storage, liquid hydrogen should be used because of its high density. Magnetic refrigeration which is based on the magnetocaloric effect of solids has the potential to achieve high thermal efficiency for hydrogen liquefaction in comparison with conventional systems using a Joule-Thomson expansion. This paper reports on recent progress on development of our magnetic refrigerator both Carnot magnetic refrigerator (CMR) and active magnetic regenerator (AMR) refrigerator.

2. Magnetic refrigeration system for hydrogen liquefaction

In order to realize a hydrogen liquefaction system using magnetic refrigeration, cascading several magnetic refrigerators is necessary to cover wide temperature ranges from heat sink temperatures to liquid hydrogen temperature (20.3 K). At this stage, the heat sink is planned as liquid natural gas (LNG) because a number of LNG plants have been built in Japan. LNG with boiling temperature at 112 K will contribute to improve the efficiency of the hydrogen magnetic refrigerator cycle[1]. In our system, there are two kinds of magnetic refrigeration cycles; CMR and AMR. Hydrogen gas is precooled to near the boiling point with an AMR refrigerator and then liquefied with a CMR[2].

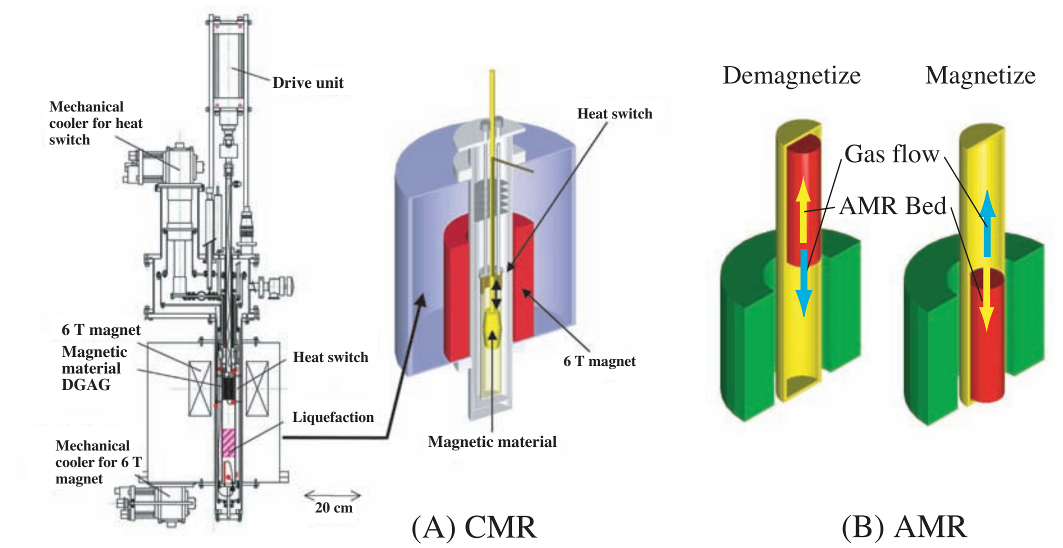

Figure 1. Cross-sectional view of the test apparatus. CMR and AMR operation are shown in (A) and (B), respectively.

3. Results and discussion

An apparatus shown in Fig. 1 has been built and tested to investigate both liquefaction process of the CMR and AMR cycle by changing the experimental conditions such as magnetic material, heat transfer gas and so on. This apparatus consists of a magnetic refrigerant, superconducting magnet with a maximum field of 6 T cooled by mechanical cooler, an electric motor that gives field change to magnetic refrigerant, and a GM cryocooler which absorbs heat exhausted by magnetic refrigerator. The details have been described elsewhere[2, 3].

3.1. Carnot Magnetic Refrigerator (CMR)

Figure 1(A) shows the schematic drawing of the CMR operation. The liquefaction principal of our magnetic refrigerator is based on the thermo-siphon method, in which liquid hydrogen is condensed directly on the surface of magnetic refrigerants and drops downward. Figure 2 shows plates made of dysprosium gadolinium aluminium garnet, $Dy_{2.4}Gd_{0.6}Al_5O_{12}$ (DGAG) that were used as magnetic refrigerant. We have successfully liquefied hydrogen gas that was precooled to a temperature slightly above the boiling point[2]. Thermal efficiency, which is determined by a ratio of cooled heat to that in an ideal Carnot demagnetization process was 50∼60 % including heat switch and condensation losses. Direct condensation on the DGAG plate gave a very high liquefaction efficiency, typically 90% in condensation process.

3.2. Active magnetic regenerator (AMR)

In high temperatures, Carnot cycle is not useful because large specific heat of magnetic material limits the adiabatic temperature change in most of the rare earth compounds to typically 1∼2 K/T. For this reason, a regenerative cycle is required to achieve a wide temperature operation with a practical field up to 6 T. An AMR separates the functions of heat pumping and temperature spanning and is considered as a combination of many cascaded magnetic refrigerators and regenerative heat exchanger. AMR refrigerators have been intensively studied in room temperature[4] and also thought to be a useful magnetic refrigerator for precooling hydrogen gas down to liquefaction temperature.

An AMR is stuffed with magnetic materials, and the regenerator matrix operates the refrigeration cycle by periodically changing magnetic field and fluid flow. It is essential for the fluid flow to be synchronized with the magnetization and demagnetization of the regenerator matrix. For example, Brayton AMR cycle consists of two isentropic (adiabatic magnetization/demagnetization) processes and two isofield (cold/hot flow) processes. We had to use simplified AMR cycle in which heat transfer gas flowed simultaneously with the magnetic material drive, since it was impossible to install independent fluid flow mechanism due to the small field space of our apparatus. Figure 1(B) shows the schematic of the simplified AMR cycle, which is described as follows. When the AMR bed is pulled up in the demagnetization process, magnetic material is demagnetized and its temperature decreased. At the same time, transfer gas, that flows downward by the motion of AMR bed, is cooled by magnetic material and the lower side of AMR becomes cold. In the magnetizing process, magnetic material is magnetized and heat transfer gas carries the magnetization heat to upward. Thus, the upper side of the AMR bed becomes hot. Consequently, temperature span will be obtained in the AMR bed.

Magnetic refrigerant of DGAG particles with 0.4 mm diameter shown in Fig. 3 was packed into a cylindrical sample holder with 40 mm in diameter and 100 mm in length. A teflon sealing ring between the sample holder and outer cylinder forced heat transfer gas to flow through the regenerator bed.

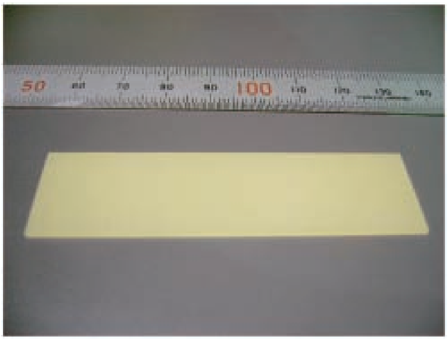

Figure 2. Photograph of a rectangular solid DGAG.

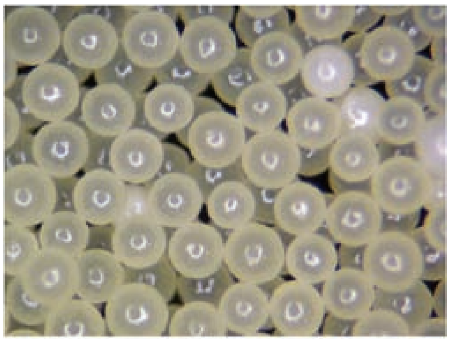

Figure 3. Photograph of spherical DGAG particles.

Figure 4 shows the experimental results on the temperature change of magnetic material at upper and lower side of regenerator bed during the AMR cycle operation. AMR bed was driven linearly with a stroke of 200 mm between the field center and weak field area by the drive motor. The total cycle period was 8 s that consisted of 2 s magnetization and demagnetization process and 2 s waiting time between every field change. Helium gas was used as heat transfer fluid at a pressure of 0.1 MPa. At the beginning of AMR operation, there was no temperature span. A temperature span was developed by driving the AMR bed and becomes steady within about 300 seconds. The obtained temperature span in AMR bed was increased with increasing magnetic field because of the increase of magnetocaloric effect. The temperature span was also increased with the increase in the pressure of transfer gas due to good heat transfer. After the AMR operation was stopped, the temperature span in the AMR bed rapidly diminished, indicating that large heat leak existed. However, AMR cycle was confirmed to expand temperature span at least two times larger than the adiabatic temperature change of DGAG around 20 K in spite of large parasitic heat leak .

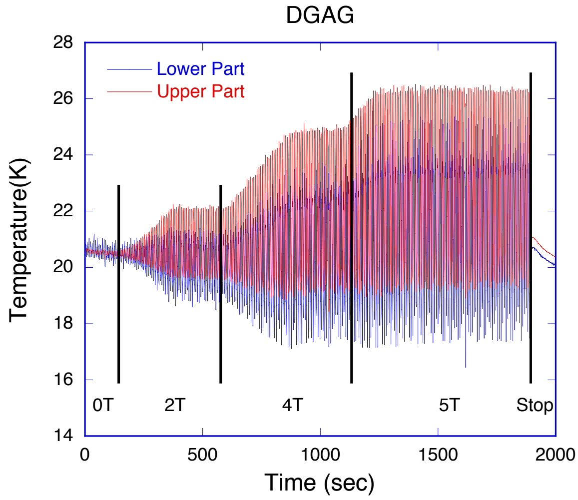

Figure 4. Test result of AMR operation. Temperature change of magnetic material at upper and lower side of the regenerator bed are shown for various magnetic field operation.

4. Summary

This paper described the recent progress on the magnetic refrigerator for hydrogen liquefaction. We have tested both CMR and AMR cycles. CMR had high liquefaction efficiency. AMR cycle was realized around 20 K for the first time. In the future, various magnetic materials such as rare earth intermetallic compounds[5, 6, 7] will be tested with this apparatus in the temperature range between 20 and 77 K.

5. Acknowledgments

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