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Magneto Caloric Properties of Polycrystalline Gd_2O_2S for an Adiabatic Demagnetization Refrigerator

H.Fukuda[1,a], Y.Kobayashi[1], R.Arai[1], K.Baba[1], H.Nakagome[1] and T. Numazawa[2]

[1]*Department of Urban Environment System, Chiba University Chiba 263-8522, Japan*
[2]*Environment and Energy Materials Division, National Institute for Materials Science Tsukuba, Ibaraki 305-0003, Japan*

Abstract. Currently, many space missions that use cryogenic equipment are being planned. In particular, high resolution sensors, such as transition edge sensors, require very low operating temperatures, below 100 mK. Adiabatic demagnetization refrigerator (ADR) systems are a useful tool for producing ultra-low temperatures in space because these devices can operate independently of gravity. The magnetic material is one of the most important components with respect to effectiveness of the cooling power. Thus, we could increase the cooling power using a magnetic material that has a large entropy change over the operating temperature range. Polycrystalline Gd_2O_2S (GOS), which was developed by Numazawa et al, can be used as such as a magnetic regenerator material. Furthermore, GOS has a very large specific heat and a magnetic phase transition temperature of about 5.2 K. These features make GOS suitable for use in the high temperature stage of an ADR. In this study, we measured and evaluated the physical properties of GOS for applications to ADRs.

1 Introduction

Many scientific measurements have been performed in microgravity environments in recent years. In the study of astrophysical phenomena the polarimetry of cosmic microwave background (CMB) is a particularly active field of research.

Detectors used in polarimetry of CMB are transition edge sensor (TES) type microcalorimeters. A TES type microcalorimeter requires an ultra-low temperature operating environment of less than 100 mK [1]. Thus, it is necessary to develop refrigerators that operate below 100 mK and can be used in space. To produce temperatures below 100 mK in space, Shirron et al. devised the continuous adiabatic demagnetization refrigerator (CADR) [2] [3]. We have also studied a continuous adiabatic demagnetization refrigerator (ADR) system consisting of four stages [4].

The ADR uses a magnetocaloric effect [5] that is caused by an entropy change of a magnetic material when the external magnetic field is changed. The entropy change of the magnetic material is described by equation (1), derived from Maxwell relations. (M : magnetization, H : magnetic field)

$$dS = \left(\frac{\partial M}{\partial T}\right)_H dH \tag{1}$$

From equation (1), the entropy change of a magnetic material can be determined by integrating the differential of the magnetization of the magnetic material in the external magnetic field. Thus, a large entropy change can be obtained if the differential of the magnetization of the magnetic material is

[a] Corresponding author: fukuda393@gmail.com

large. Thus, magnetic materials that have large entropy changes in desired temperature range could be used to increase the cooling power of ADRs.

Polycrystalline Gd_2O_2S (GOS), which was developed by Numazawa et al [6] [7], be used as such a magnetic regenerator material. Moreover, GOS is suitable for use in the high temperature stage of an ADR because its magnetic phase transition temperature is about 5.2 K. In this study, we measured the magnetization and specific heat of GOS and evaluated the physical properties of GOS for application to ADRs.

2 Polycrystalline Gd2O2S

GOS is an antiferromagnetic material that has its spins ordered in an antiparallel arrangement. Figure 1 illustrates an ordered arrangement of ferromagnetic and antiferromagnetic spins. A ferromagnetic material has a spontaneous magnetic moment that exists even if no magnetic field is applied. Antiferromagnetic materials do not have a spontaneous magnetic moment because their spins are ordered in an antiparallel arrangement. Moreover, antiferromagnetic materials generally do not feature strong magnetization when magnetized by external magnetic fields because their spins cancel each other. However, when the external magnetic field becomes strong the spins become aligned in the same direction as the external magnetic field. This phenomenon is called spin-flop and it produces a considerable magnetization change when all the spins become aligned in the same direction. If spin-flop is applied to an ADR, it is possible to induce large temperature changes because the large magnetization change leads to a large entropy change. GOS has been put to practical use as a magnetic regenerator material because it typically shows a λ phase transition and has a huge specific heat.

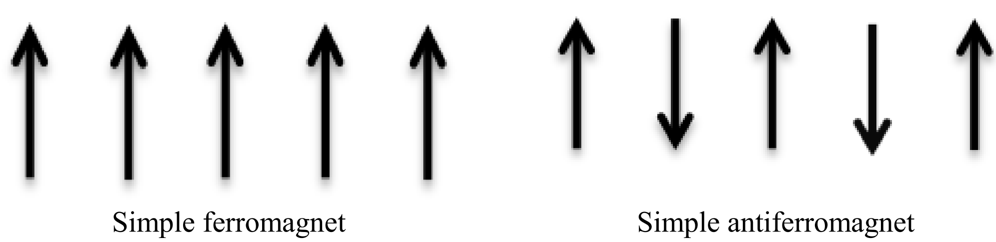

Figure 1. Ordered arrangement of spins

3 Experimental results

We measured the magnetization of GOS using a magnetic property measurement system (MPMS, Quantum Design) and the specific heat of GOS using a physical property measurement system (PPMS, Quantum Design). Figure 2 shows the sample of GOS used in measurement. The magnetization of GOS in an external magnetic field was measured at 4, 5, and 6 K when magnetized up to 5 T. The variation of GOS magnetization with temperature was measured at 0.1, 3, and 5 T for a temperature increase from 2.2 to 15 K. The change in specific heat with temperature was measured at 0, and 5 T for an increase from 2.2 to 15 K. The variation of GOS magnetization in an external magnetic field, the variation of GOS magnetization with temperature, and the specific heat with temperature are shown in Figures 3–5, respectively.

A spin–flop occurring in these systems should appear as a large magnetization change. However, Figure 3 shows that the magnetization of GOS increased linearly indicating that no spin-flop occurred. One reason for this is that GOS requires a magnetization greater than 5 T to reverse its spins.

Next, we focus on Figure 5, the specific heat with temperature. From this result, we determined the phase transition of GOS to be around 5.2 K. In addition, we calculated the entropy change of GOS using equation (2), as show in Figure 6, and interpolated the entropy using external interpolation.

$$S = \int_0^T \frac{C}{T} dT \qquad (2)$$

Figure 6 shows that there was no effective entropy change below 4 K, which is the high temperature stage of an ADR. The entropy change of GOS was less than that of $GdLiF_4$, which is currently used in the high temperature stage of ADRs [8]. However, when we see figure 7 and 8, we could confirm that the entropy curve at 0 T was larger than that at 5 T, between 5 and 6 K. This is considered an inverse magnetocaloric effect [9] [10] [11]. The entropy of GOS and the entropy of GOS between 5 and 6 K are shown in Figure 7 and 8.

Figure 2. Sample of GOS used in measurement

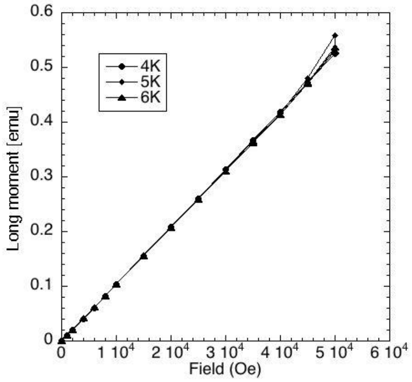

Figure 3. Variation of Gd2O2S magnetization with external magnetic field at 4, 5, and 6 K when magnetized to 5 T.

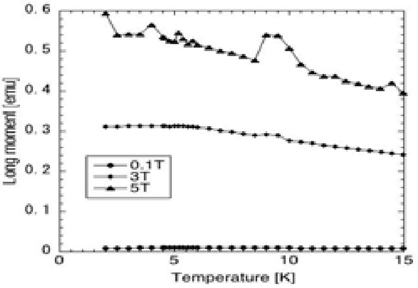

Figure 4. Variation of Gd2O2S magnetization with temperature at 0.1, 3, and 5 T as temperature was increased from 2 to 15 K.

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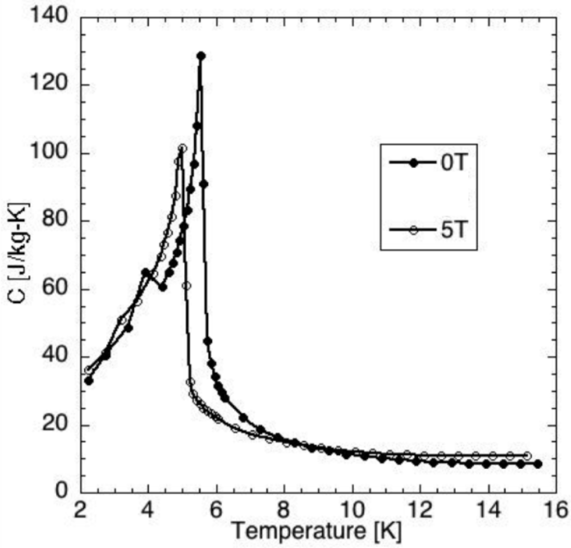

Figure 5. Specific heat with temperature at 0, and 5 T when temperature was increased from 2.2 to 15 K .

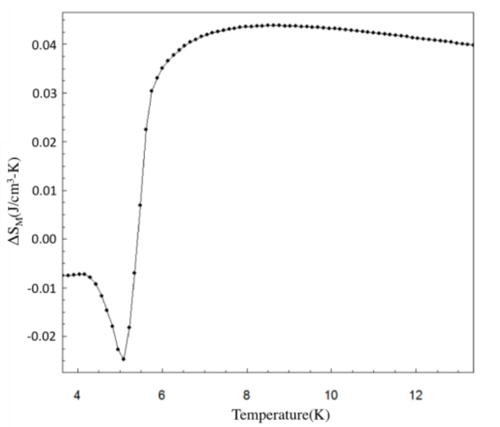

Figure 6. Entropy change of Gd_2O_2S as functions of magnetic field and temperature.

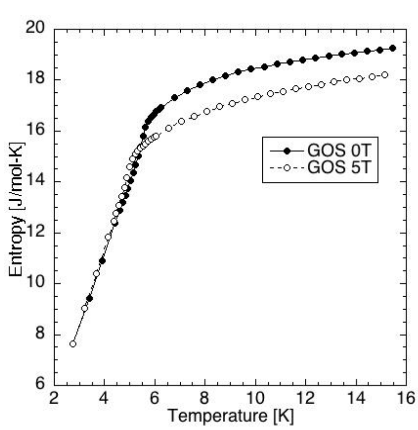

Figure 7. Entropy of Gd2O2S as functions of magnetic field and temperature.

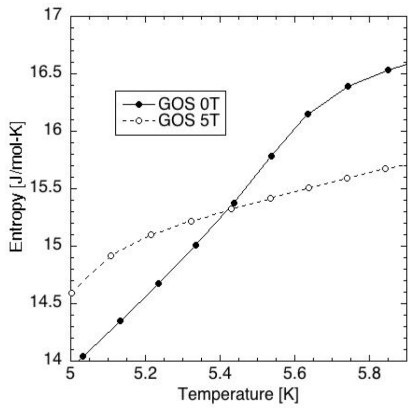

Figure 8. Entropy of Gd2O2S as functions of magnetic field and temperature between 5 and 6 K.

4 Conclusions

We measured the variation of GOS magnetization with external magnetization and temperature. However, we did not observe a spin-flop because and no notable magnetization change occurred when GOS was magnetized to 5 T. Therefore, we assume that a large external magnetic field, for example 10 T is required to observe spin flop behavior.

We measured the variation of the specific heat of GOS with temperature and calculated the entropy change of GOS. Although we could not find an efficient entropy change around 4 K, which is the high temperature stage of an ADR, we confirmed inverse magnetocaloric effects, as shown in Figure 8. Thus, in our future work we will measure the entropy properties of GOS in a high external magnetic field.

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