Cooling Performance of Multilayer Ceramic Regenerator Materials

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ABSTRACT

Ceramic regenerator particles with an average diameter of 0.25 mm have been successfully fabricated using a two-layer structure of magnetic materials: the outer shell layer being Tb_2O_2S, and the inner core being $(Gd_{0.1}Tb_{0.9})_2O_2S$. The measured heat capacity data show twin peaks relating to the two component materials, $(Gd_{0.1}Tb_{0.9})_2O_2S$ and Tb_2O_2S, and the entire curve has become broader and wider. The mechanical properties of strength and hardness of the two-layer ceramic material are similar to those of other ceramic regenerator materials such as GOS. Cooling performance comparison tests have been conducted using the two-layer ceramic material in comparison with $HoCu_2$ and GOS in two kinds of GM cryocoolers: 1.0 W and 0.1 W at 4.2 K. The experimental data for the 1W GM show that the 4 K cooling capacity using the two-layer material was not higher than with the $HoCu_2$ or $HoCu_2$+GOS regenerator configurations. However, in the case of the 0.1W GM, the 4 K cooling capacity of the two-layer regenerator material was higher than with $HoCu_2$ alone, and its usability was confirmed.

INTRODUCTION

Over the past several years we have studied oxide magnetic materials for use in regenerators of 4 K cryocoolers. In particular, GAP = $GdAlO_3$ and GOS = Gd_2O_2S, have demonstrated improvement of the cooling performance of 4 K cryocoolers.[1,2] In order to expand the usability of ceramic regenerator materials, the $(Gd_xTb_{1-x})_2O_2S$ system has been proposed as a candidate material to add additional heat capacity to regenerators in the temperature range between 6 K and 10 K.[3] An advantage of the $(Gd_xTb_{1-x})_2O_2S$ system is that it offers the variable x that allows its heat-capacity peak to be tailored to a particular temperature. The possibility also exists to generate a multilayer regenerator material with a tailorable heat capacity based on two or more layers of $(Gd_xTb_{1-x})_2O_2S$, each with a different value of x. In this work we explore the fabrication and thermal performance of a two-layer system involving materials with x = 0 and 0.1, i.e. Tb_2O_2S and $(Gd_{0.1}Tb_{0.9})_2O_2S$. Data are presented on the properties of this two-layer magnetic material as well as its cooling performance in 4 K GM cryocoolers.

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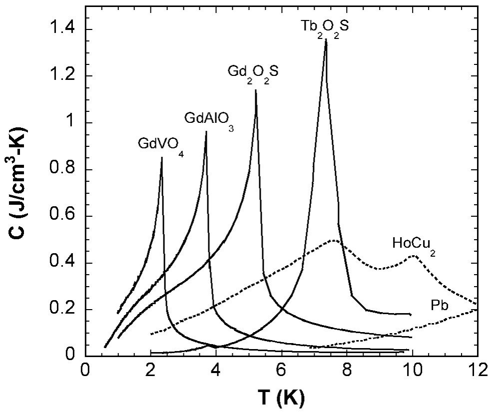

Figure 1. Heat capacity data at zero magnetic field for the oxide magnetic materials $GdVO_4$, $GdAlO_3$, Gd_2O_2S and Tb_2O_2S; also shown are Pb and $HoCu_2$.

CERAMIC REGENERATOR MATERIALS

Ceramic oxide magnetic materials show excellent properties as regenerator materials in 4 K cryocoolers. These properties include high heat capacity per unit volume, easy fabrication of spherical particles, high thermal conductivity, high mechanical strength, and weak dependence on magnetic field.[1,2] Figure 1 shows heat capacity data in the 2 K to 10 K temperature range for the oxide magnetic materials $GdVO_4$, $GdAlO_3$, Gd_2O_2S and Tb_2O_2S. These four kinds of oxide materials can be used as high-heat-capacity materials for regenerators or as thermal anchors. These materials have a typical lambda type anomaly coming from an antiferromagnetic transition. Since this kind of anomaly has a sharp peak with a narrow half width of heat capacity, two or three materials have to be used to cover the temperature range below 10 K.

MULTILAYER CERAMIC REGENERATOR MATERIAL

In order to provide a high heat capacity for the 6 K region, the $(Gd_xTb_{1-x})_2O_2S$ system has been considered.[3] Figure 2 shows the heat capacity of fabricated bulk samples of $(Gd_xTb_{1-x})_2O_2S$ for various values of the composition x between zero and one. Note that samples for $x \leq 0.1$ have their peak heat capacity at temperatures between 6 K and 8 K where the heat capacity of GOS ($x = 1$) and Pb is very low. Since the heat capacity peak of the $(Gd_xTb_{1-x})_2O_2S$ system is so sharp, it is desirable

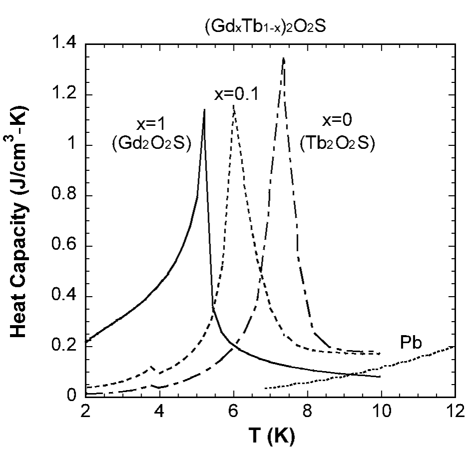

Figure 2. Volumetric heat capacity of the $(Gd_xTb_{1-x})_2O_2S$ system.

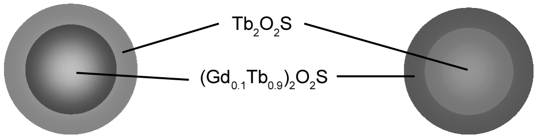

Figure 3. Structure of two-layer magnetic regenerator material. The particle consists of Tb_2O_2S and $(Gd_{0.1}Tb_{0.9})_2O_2S$ for inner and outer layer in case (a), or outer and inner layer in case (b), respectively.

to combine several $(Gd_xTb_{1-x})_2O_2S$ materials with different values of x to extend the heat capacity over a broader range of temperatures.

Multilayer particles having different peak temperatures have been successfully made for this purpose.[4] To work as a regenerator material, these layers have to be firmly connected mechanically and thermally, while they are magnetically independent. A two-layer system consisting of $(Gd_{0.1}Tb_{0.9})_2O_2S$ for the inner layer and Tb_2O_2S for the outer layer has been made to provide high heat capacity between 6 K and 8 K as schematically shown in Fig. 3(a). In this case, the volumetric ratio of inner material to outer material is 1 to 1.

Figure 4 shows experimental results for the heat capacity of the two layer $(Gd_{0.1}Tb_{0.9})_2O_2S$+ Tb_2O_2S sample as well as for monolayer $(Gd_{0.1}Tb_{0.9})_2O_2S$ and Tb_2O_2S samples. All samples are real regenerator particles with an average diameter of 0.25 mm. The results for the two-layer material clearly indicates the superposition of two magnetic elements. The heat capacity curve has twin peaks relating to those of the $(Gd_{0.1}Tb_{0.9})_2O_2S$ and Tb_2O_2S magnetic elements, and the entire curve becomes broader and wider. The mechanical properties of strength and hardness of the two-layer ceramic material are the same as other ceramics regenerator materials. Thus, it is concluded that the multilayer structure can be used to usefully tailor the heat capacity of the regenerator particles.

We also considered a different structure for the two-layer particle as shown in Fig. 3(b). In this case, the two-layer structure is opposite to the case of Fig. 3(a); i.e., the inner and outer layers consist of Tb_2O_2S and $(Gd_{0.1}Tb_{0.9})_2O_2S$, respectively. The peak temperature of the heat capacity of Tb_2O_2S is higher than that of $(Gd_{0.1}Tb_{0.9})_2O_2S$ as shown in Fig. 2. This is somewhat interesting when we consider the thermal penetration depth of the two-layer regenerator particle. If the thickness of the outer layer of the two-layer particle is larger than the thermal penetration depth, the heat transfer to the inner layer will be insufficient. We have tried to make the structure of Fig. 3(b), though, we have not yet successfully completed the fabrication. The main difficulty is the difference of the heat treatment temperature of the two materials. In general, the heat treatment temperature of the outer layer must be higher than that of the inner layer because HIP (Hot Isostatic Processing) is used as the final fabrication stage.[4] The heat treatment temperature of Tb_2O_2S is higher than that of $(Gd_{0.1}Tb_{0.9})_2O_2S$, and this condition is inconsistent with the structure in Fig. 3(b).

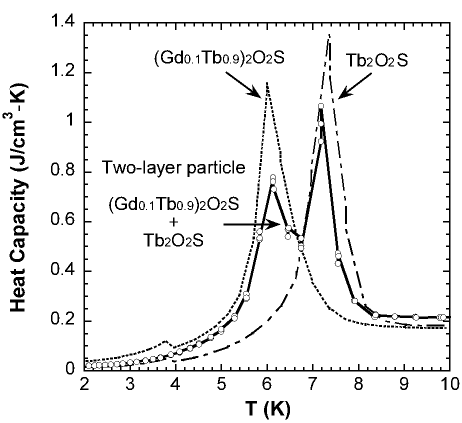

Figure 4. Experimental results on the heat capacity for $(Gd_{0.1}Tb_{0.9})_2O_2S$, Tb_2O_2S and two-layer $(Gd_{0.1}Tb_{0.9})_2O_2S+Tb_2O_2S$ regenerator particles.

Table 1. Configurations of 2nd regenerator for 1 W GM cryocooler.

Configuration #	Pb (g)	$HoCu_2$ (g)	Two-layer GTOS (g)	GOS (g)
1	469	191	0	0
2	469	0	142	0
3	469	0	104	40
4	469	0	77	60
5	469	0	54	84
6	469	0	35	101

Table 2. Configurations of 2nd regenerator for 0.1 W G-M cryocooler.

Configuration #	Pb (g)	$HoCu_2$ (g)	Two-layer GTOS (g)	GOS (g)
1	74	73	0	0
2	74	0	56	0
3	74	0	42	14
4	74	0	30	28
5	74	0	15	43
6	74	0	0	57

RESULTS AND DISCUSSION

In order to test the cooling performance of the two-layer regenerator material, two types of 4 K GM cycle cryocoolers were chosen: a 1W at 4.2 K (Sumitomo SRDK-408D with a compressor unit of 7.5 kW) and a 0.1 W at 4.2 K (Sumitomo SRDK-101D with a compressor unit of 1.3 kW). Several second stage regenerator configurations were investigated as shown in Tables 1 and 2 for the 1 W type and 0.1 W GM coolers, respectively. In these tests, the weight of Pb was fixed, and two kinds of configurations were tested: 1) Pb+$HoCu_2$, and 2) Pb+GTOS+GOS, where GTOS is the two-layer $(Gd_{0.1}Tb_{0.9})_2O_2S$+Tb_2O_2S regenerator material.

1W 4 K GM Cryocooler

Figure 5 shows the cooling capacity of the 2nd stage of the 1W GM cryocooler at 4.2 K as a function of 2nd-stage regenerator configuration and 1st-stage temperature. Configuration #1 (Pb+$HoCu_2$) is the configuration of the 2nd-stage regenerator commonly used in the commercial 4 K GM cryocooler where the cooling capacity is typically about 1 W at 4.2 K. For configuration #2, the $HoCu_2$ in the 2nd-stage regenerator is entirely replaced by the two-layer ceramic material GTOS. The cooling capacity decreased significantly down to 0.2 W at 4.2 K. This is because the GTOS does not provide sufficiently high heat capacity at 4.2 K. In configurations from #3 to #6,

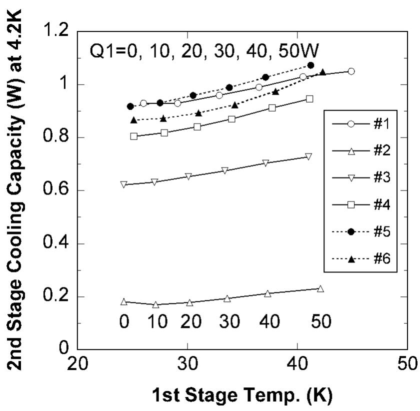

Figure 5. Experimental results for 2nd-stage cooling capacity at 4.2 K for the 1W GM cryocooler as a function of 2nd-stage-regenerator configuration and 1st-stage temperature.

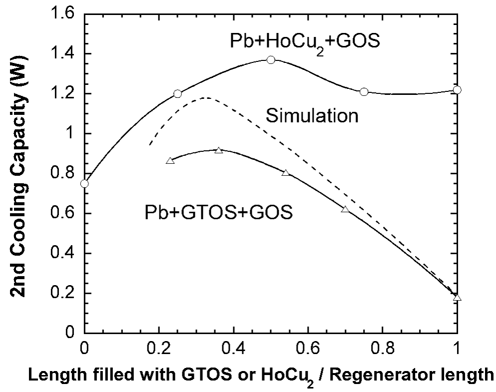

Figure 6. Experimental results for the cooling capacity of the 2nd stage at 4.2 K as a function of the fraction of the second-stage regenerator length filled with $HoCu_2$+GOS or GTOS+GOS.

GOS material was added to compensate for the lack of heat capacity of the GTOS material in the 4 K region. There are clear improvements by adding the GOS to the GTOS. In fact, configuration #5 has a slightly higher cooling capacity than that of configuration #1 (Pb+$HoCu_2$).

Figure 6 shows cooling capacity curves at 4.2 K for two 2nd-stage regenerator configurations with no heat load on the 1st stage. This gives a much clearer view for the regenerator configurations. There is an optimum volume ratio between the GTOS and the GOS, and a similar optimum ratio of $HoCu_2$ to GOS for that configuration. The optimum 2nd-stage regenerator configuration of Pb+$HoCu_2$+GOS gives a higher cooling capacity than that of the Pb+GTOS+GOS configuration.

In order to confirm the trends shown by the experimental data for the two regenerator configuration, a numerical simulation was carried out based on a Nodal analysis. The dotted line in Fig. 6 shows the calculated results for the Pb+GTOS+GOS configuration. The experimental data are modestly lower than the simulation curve, especially around the peak temperature. We have not identified the cause of this difference, but further optimization of the regenerator may increase the cooling capacity in comparison to the $HoCu_2$+GOS configuration.

0.1W 4 K GM Cryocooler

Similar cooling tests were carried out with the 0.1W GM cryocooler. Figure 7 shows the cooling capacity of the 2nd stage at 4.2 K as a function of 2nd-stage-regenerator configuration and 1st-stage temperature.. Adding GOS to the GTOS leads to a clear improvement.

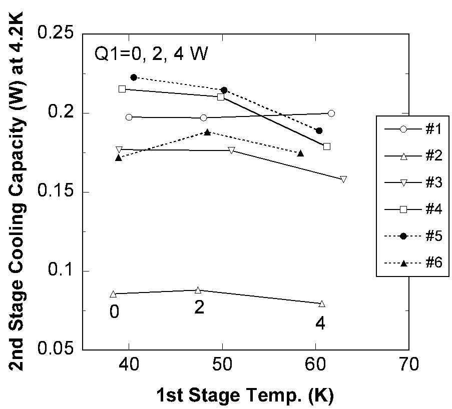

Figure 7. Experimental results for 2nd-stage cooling capacity at 4.2 K for the 0.1 W GM cryocooler as a function of 2nd-stage-regenerator configuration and 1st-stage temperature.

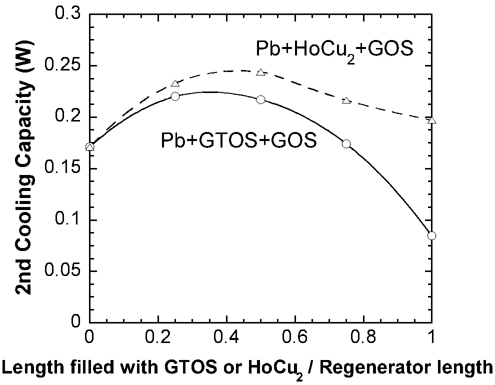

Figure 8. Experimental results on the cooling capacity of 2nd stage regenerator at 4.2 K for the 1st stage temperature for 0.1 W GM cryocooler.

Figure 8 shows cooling capacity curves at 4.2 K for various 2nd-stage regenerator configurations with no heat load on the 1st stage. The dotted line shows the cooling capacity data previously obtained with the Pb+$HoCu_2$+GOS configuration. This configuration is higher than that of the Pb+GTOS+GOS one (solid line). However, the difference is relatively small compared with the case of the 1W GM cryocooler. Also, the Pb+GTOS+GOS configuration gives higher cooling capacity than that of Pb+$HoCu_2$ configuration. Thus, we conclude that the two-layer GTOS regenerator material may be more effective for lower cooling capacity GM cryocoolers.

CONCLUSIONS

The concept of a multilayer regenerator material, with its tailorable heat capacity, provides flexibility in the design of regenerator configurations. Conclusions from fabricating the GTOS material and testing it in two-stage GM cryocoolers include:

- Thermal penetration depth must be carefully considered for this kind of regenerator particle
- The heat treatment temperature for each layer is important to the fabrication process of the particles. More precise control of the heat treatment process is needed.
- A mixture of the two-layer ceramic regenerator material GTOS with GOS gives better cooling performance than that of $HoCu_2$ alone. However, this mostly comes from the effect of the GOS. Simulation results suggest that higher cooling capacity may be obtainable with the two-layer GTOS+GOS by further tuning of the regenerator configuration in the 1W GM cooler.

REFERENCES

1. Numazawa, T., Kamiya, K., Satoh, T., Nozawa, H. and Yanagitani, T., "A New Ceramic Magnetic Regenerator Material for 4 K Cryocoolers," *Cryocoolers 12*, Kluwer Academic/Plenum Publishers, New York (2002), p. 473.

2. Numazawa, T., Kamiya, K., Satoh, T., Nozawa, H. and Yanagitani, T., "Status of the Development of Ceramic Regenerator Materials," *Cryocoolers 13*, Kluwer Academic/Plenum Publishers, New York (2004), p. 373.

3. Numazawa, T., Kamiya, K., Satoh, T., Nozawa, H. and Yanagitani, T., "Performance of Gd-Tb Oxysulfide Ceramic Regenerator Material for GM Cryocoolers," *Adv. in Cryogenic Engineering*, Vol. 49 (2004), p. 1958.

4. Numazawa, T., Kamiya, K., Satoh, T., Nozawa, H. and Yanagitani, T., "Multilayer Ceramic Regenerator Materials for 4 K Cooling," *Adv. in Cryogenic Engineering*, Vol. 51B (2006), p. 1904.