Size and Composition Optimization of $Er_xHo_{1-x}N$ Regenerators for 4K-GM Cryocoolers

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ABSTRACT

$Er_xHo_{1-x}N$ spheres were synthesized by nitriding Er_xHo_{1-x} metallic spheres with different sizes in the hot isostatic pressing method in a nitrogen sphere (1550°C and 195 MPa of 99.9999 % nitrogen). Cooling tests were performed using a commercially available two-stage GM cryocooler (Model SRDK-101D, Sumitomo Heavy Industries Ltd.) by loading $Er_xHo_{1-x}N$ spheres in the second stage regenerator column instead of $HoCu_2$. The optimal size of $Er_xHo_{1-x}N$ spheres was found to be approximately 150-200 µm. The most suitable composition was $Er_{0.75}Ho_{0.25}N$. When $Er_{0.75}Ho_{0.25}N$ with a size of 150-180 µm was used, the cooling power of the GM cryocooler at 4.2 K reached 0.266 W.

INTRODUCTION

Gifford-McMahon (GM) cryocoolers are cryogenic refrigerators widely used for helium and hydrogen liquefaction, and for systems using superconducting magnets such as magnetic resonance imaging diagnosis devices and magnetic levitation trains. Their performance and efficiency depend on the regenerator materials used. The most important requirement for the regenerator materials are high specific heat around the working temperatures. To improve the efficiency, the researchers and engineers are seeking advanced regenerator materials[1-4].

ErN and HoN are ferromagnetic with transition temperatures of 4 and 12 K, respectively[5, 6]. They show large magnetic specific heat around their transition temperatures[5,7]. Nakano et al.[8] reported that they perform well as regenerators for 4K-GM cryocooler and the cooling power (CP) of the GM cryocooler at 4.2 K became high when the rare earth nitride spheres are a smaller size[8]. However, using nitride spheres smaller than 180 µm was not studied, so the optimal size of the nitride regenerator spheres has still been unknown.

Binary nitrides, $Er_xHo_{1-x}N$, are also promising regenerator materials because the peak temperature of the specific heat can be adjusted in the rage of 4 to 12 K by controlling the composition x[9]. Shoda et al.[10] measured the CP of the GM cryocooler using $Er_xHo_{1-x}N$ (x = 0.5, 0.75 and 1) regenerators. When $Er_{0.75}Ho_{0.25}N$ with a size of 180-212 µm was used, the CP of the GM cryocooler at 4.2 K (nominal CP at 4.2 K is 0.1 W) reached 0.206 W. However, the size and filling ratio of $Er_xHo_{1-x}N$ spheres to the second regenerator column were not unified in previous reports. The optimal composition of $Er_xHo_{1-x}N$ has not been clear.

The aim of this work is to find the most suitable composition and sphere diameter of $Er_xHo_{1-x}N$ as a regenerator material for a 4K GM cryocooler.

EXPERIMENTAL

$Er_xHo_{1-x}N$ (x = 0.75 and 0.5) spheres were synthesized by nitriding Er_xHo_{1-x} metallic spheres with different sizes in the hot isostatic pressing method in a nitrogen sphere (1550°C and 195 MPa of 99.9999 % nitrogen). Formation of $Er_xHo_{1-x}N$ was confirmed by the X-ray diffraction. The morphology of $Er_xHo_{1-x}N$ was observed by a scanning electron microscope (SEM). Cooling tests were performed using a commercially available two-stage GM cryocooler (Sumitomo Heavy Industries Ltd. Model SRDK-101D) with nominal cooling power of 0.1 W at 4.2 K. The operating conditions were the standard ones provided by supplier. The second regenerator column consists of packed Pb spheres, 200 μm diameter, together with the cryogenic magnetic regenerator. The initial regenerator material of $HoCu_2$ (180-300 μm in diameter) was replaced $Er_xHo_{1-x}N$ spheres with different sizes. The weights of the $Er_xHo_{1-x}N$ regenerator materials loaded in the column were measured. By varying the input power to a heater attached to the cold head and monitoring the equilibrium temperature, T, cooling power (CP) versus T curves were obtained. The lowest achieving temperature was determined when CP = 0. Size range, average size, filling amount and filling space ratio (7.32 cm^3 corresponds to 100 %) of $Er_xHo_{1-x}N$ spheres loaded to the second regenerator column are summarized in Table 1.

RESULTS AND DISCUSSION

The SEM images of $Er_{0.5}Ho_{0.5}N$ with different sizes are shown in Fig. 1. The figure indicates that the spherical $Er_{0.5}Ho_{0.5}N$, which was suitable for regenerators of cryocoolers, was obtained. Figure 2

Table 1. Composition, size range, average size, filling amount and filling space ratio of regenerator materials used in cooling tests of 4K-GM cryocooler.

Composition	Size range (μm)	Ave. size (μm)	Filling amount (g)	Filling space ratio (%)
$Er_{0.5}Ho_{0.5}N$	75-150	113	40	52
	180-212	196	41	54
	250-300	275	36	47
$Er_{0.75}Ho_{0.25}N$	150-180	175	40	52
	180-212	196	40	52
ErN [8]	180-212	196	41	53
	212-250	231	42	54
	250-300	275	41	54
$HoCu_2$	150-300	225	40	62

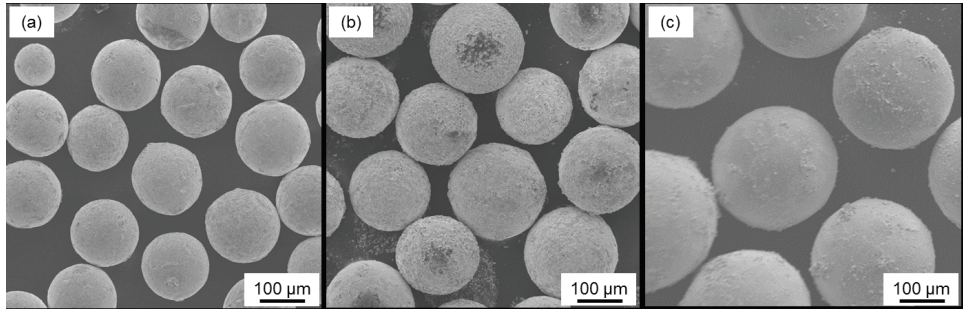**Figure 1.** SEM images of Er0.5Ho0.5N regenerators with size of (a)75-150 mm, (b) 180-212 mm, (c) 250-300 mm.

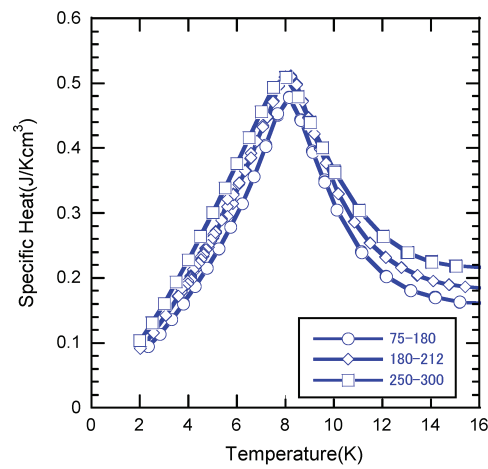**Figure 2.** Temperature dependence of specific heats of Er0.5Ho0.5N with different sizes.

gives the temperature dependences of specific heat of Er$_{0.5}$Ho$_{0.5}$N with different sizes. Regardless of the size, almost the same temperature dependence of the specific heat (peak temperature and amplitude) was obtained. No size dependency of the specific heat of the other compositions of Er$_x$Ho$_{1-x}$N was also observed.

Figure 3 shows the temperature dependence of CP of the GM cryocooler using Er$_{0.5}$Ho$_{0.5}$N with different sizes as regenerators. This figure means that Er$_{0.5}$Ho$_{0.5}$N, regardless of the sizes, functions as regenerator materials of GM cryocooler which reaches 4.2 K. The CPs of the cryocooler at a temperature were changed when the different size of Er$_{0.5}$Ho$_{0.5}$N regenerators were used. The 180-212 μm is the optimal size for Er$_{0.5}$Ho$_{0.5}$N to obtain the highest CP at 4.2 K.

Figure 4 indicates the temperature dependence of CP for the cryocooler using Er$_{0.75}$Ho$_{0.25}$N. Higher CPs were observed compared to the Er$_{0.5}$Ho$_{0.5}$N regenerators. The CPs of the cryocooler at different temperature using HoCu$_2$, which is employed in regenerators of commercially available

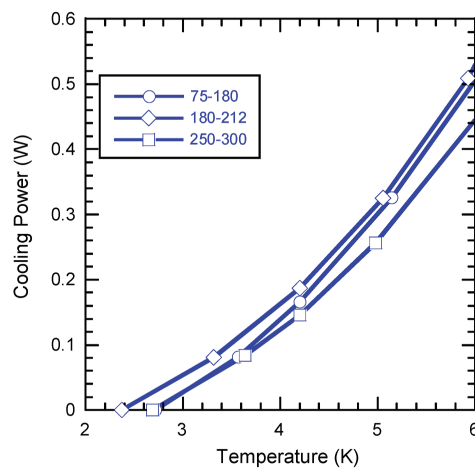**Figure 3.** Cooling power of GM cryocooler using different size of Er0.5Ho0.5N regenerators.

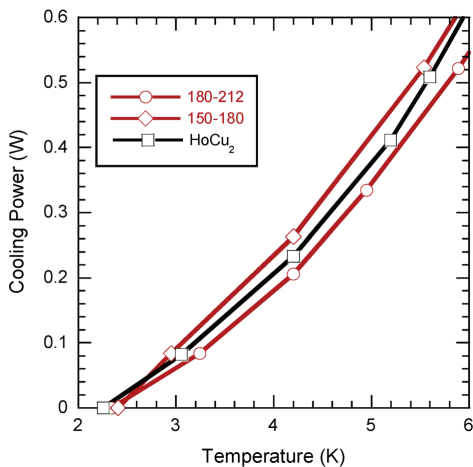**Figure 4.** Cooling power of GM cryocooler using Er0.75Ho0.25N regenerators. The results of HoCu2 regenerator is also displayed for comparison.

4K GM cryocoolers, are also given in this figure. At a glance, the CP of $Er_{0.75}Ho_{0.25}N$ with 150-180 μm in diameter exceeds that of $HoCu_2$ (CP at 4.2 K is 0.233 W) can be seen.

Figure 5 shows the plots of the average sphere size vs. CP at 4.2 K for different compositions of $Er_xHo_{1-x}N$ (x = 1, 0.75 and 0.5). The data for ErN are previously reported[8]. The optimal size of $Er_xHo_{1-x}N$ spheres was found to be approximately 150-200 μm. When compared to the sphere diameter of 180-212 mm, which has a complete set of data by all composition, the composition $x = 0.75$ shows the highest CP. Therefore, the most suitable composition of $Er_xHo_{1-x}N$ is $x = 0.75$ as regenerator for 4K GM cryocooler. As shown in Table 1, filling space ratios of $Er_xHo_{1-x}N$ regenerators are less than that of $HoCu_2$. It is thought that the lower filling space ratios of $Er_xHo_{1-x}N$ are due to rough surface of them as seen in Figure 1. The filling space ratios of $Er_xHo_{1-x}N$ spheres will be improved by surface treatments. From the results of calculation by REGEN 3.3 code [11], CP is proportional to
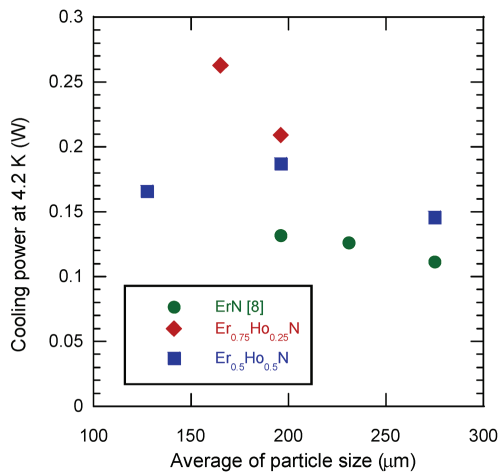

Figure 5. Average sphere size vs. CP at 4.2 K plots for the different composition of ErxHo1-xN (x = 1, 0.75 and 0.5)

the filling space ratio in the range from 50 to 70% [10]. If $Er_{0.75}Ho_{0.25}N$ with the size of 150-180 mm is filled by the same filling space ratio as $HoCu_2$, the CP of 4K GM cryocoolers will be increased to 0.317 W at 4.2 K.

CONCLUSIONS

To obtain the most suitable composition and the optimal sphere diameter of $Er_xHo_{1-x}N$ as regenerator materials for 4K GM cryocoolers, various size of $Er_xHo_{1-x}N$ (x = 1, 0.75 and 0.5) were prepared. $Er_xHo_{1-x}N$ sphere regenerators with different size were loaded into second regenerator column of 4K GM cryocooler with nominal cooling power of 0.1 W at 4.2 K. The temperature dependences of the cooling power were measured. It was found that 150-200 μm is the optimal size for $Er_xHo_{1-x}N$ spheres as regenerators of 4K-GM cryocoolers. Composition of the $Er_xHo_{1-x}N$ by which the highest cooling was obtained was x = 0.75. Using $Er_{0.75}Ho_{0.25}N$ with the size of 150-180 μm as regenerator, the cooling power of the GM cryocooler at 4.2 K reached 0.266 W. This value is higher than that in a case using $HoCu_2$.

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