[Article ID] 1003- 6326(2002)02- 0294- 03

Structure and magnetic properties of high-performanced $Sm_2(Co, Fe, Cu, Zr)_{17}$ alloys①

LI Li-ya(李丽娅), YI Jian-hong(易健宏), PENG Yuan-dong(彭元东), TANG Xin-wen(唐新文), ZENG Qing-ling(曾庆灵), FU Ying-sheng(付应生), LUO Shu-dong(罗述东)
(State Key Laboratory for Powder Metallurgy, Central South University, Changsha 410083, China)

[Abstract] The magnetic properties of $Sm_2(Co, Fe, Cu, Zr)_{17}$ alloys are very sensitive to heat treatments. A magnet with B_r of 1.08 T, H_{ci} of 2 312 kA·m^{-1} and $(BH)_{max}$ of 211.0 kJ·m^{-3} has been obtained after sintering at 1 201 ℃ and aging at 830 ℃. The magnet has a very low temperature coefficient (β) of − 0.18%/℃. The maximum operating temperature is dramatically higher than commercial magnets. The microstructure analysis shows that $Sm_2(Co, Fe)_{17}$, $Sm(Co, Cu)_5$ and Co_2Zr are found in the magnet. The EDAX analysis shows that the cells are enriched in Fe, but depleted in Cu and Zr, whereas the cell boundaries are Cu and Zr enriched, but Fe depleted.

[Key words] magnets; temperature coefficient; magnetic and microstructure properties
[CLC number] TM 273 **[Document code]** A

1 INTRODUCTION

The high Curie temperature and lowest temperature coefficient of the Sm_2Co_{17} permanent magnets make them be ideal candidates for high temperature applications[1]. Studies have demonstrated that the high temperature performance of the magnet alloys can be significantly improved by applying higher Sm, Cu, Zr content, and lower Fe content. This results in high intrinsic coercivity and low temperature coefficient of the magnet alloys at high temperature[2]. However, these approaches all lead to decreased energy product [$(BH)_{max}$]. Other studies show that '1: 7' type Sm-Co alloys have very low temperature coefficient and can be used up to 500 ℃, but their properties at room temperature are decreased[3,4].

The temperature coefficient is the most important factor in the development of high temperature magnets. The temperature coefficient of coercivity (β) between room temperature (T_0) and operating temperature (T_1) is defined as[1]

$$\beta = \{[H_{ci}(T_0) - H_{ci}(T_1)]/[H_{ci}(T_0) \times (T_0 - T_1)]\} \times 100\% \quad (1)$$

The maximum operating temperature (MOT) can be expressed as[4]

$$MOT = T_0 + \{[H_{ci}(T_0) - 400]/[H_{ci}(T_0) \times (-\beta)]\} \times 100\% \quad (2)$$

According to Eqn.(2), the high temperature permanent magnets are possible when the β is low and the room temperature coercivity is moderately high.

In this paper, we developed a high temperature Sm_2Co_{17} magnet based on the former experiments, and studied the properties at room temperature and high temperature.

2 EXPERIMENTAL

Alloys of compositions 25.5% Sm, 49.5% Co, 18% Fe, 4% Cu, 3% Zr were copper melted in a high purity argon atmosphere with an excess of samarium in order to compensate the samarium loss during melting. Alloy ingots were ball milled and then compacted in a magnetic field. The green compacts were sintered at 1 196, 1 201 and 1 206 ℃, respectively, and then subjected to a solution thermal treatment. The solutionized samples were aged at 830 ℃ for 20 h, followed by a slow cooling at 0.5 ℃/min to 370 ℃ for 12 h, and then quenched to room temperature. For the convenience of discussion, these three alloys are designed as alloys A, B and C, respectively. Magnetic properties were measured using a vibrating sample magnetometer with a maximum applied field of 30 kOe. Microstructure analysis was carried out using a scanning electron microscopy, X-ray diffraction and EDAX. The irreversible flux loss was estimated by measuring the flux-difference with a Helmholtz coil before and after exposing the magnet to elevated temperatures for 7 h in an argon environment.

3 RESULTS AND DISCUSSION

3.1 Magnetic properties

Table 1 shows the magnetic properties of magnets at room temperature and 200 ℃ included in this paper. It can be seen that sample B has the best

① **[Foundation item]** Project supported by the Foundation for University Key Teacher by the Ministry of Educatation
[Received date] 2001- 05- 21; **[Accepted date]** 2001- 11- 21

Table 1 Magnetic properties at 20 ℃ and 200 ℃ for magnets included in this study

Sample	20 ℃			200 ℃		
	B_r/T	H_{ci}/(kA·m^{-1})	$(BH)_{max}$/(kJ·m^{-3})	B_r/T	H_{ci}/(kA·m^{-1})	$(BH)_{max}$/(kJ·m^{-3})
A	0.99	1904.8	181.5	0.91	1469.6	149.0
B	1.08	2312.0	211.0	0.99	1501.6	181.4
C	0.99	1810.4	180.6	0.91	1348.0	148.0

magnetic properties of the three magnets. With the sintering temperature raising from 1196 ℃ to 1206 ℃, its B_r varies between 0.99T and 1.08T, and $(BH)_{max}$ varies between 181.5kJ/m^3 and 211.0kJ/m^3. Since the three magnets are of the same chemical composition, the wide variations show that the properties of these magnets are very sensitive to the sintering temperature.

The experiment results show that sample *B* has an H_{ci} of 1501.6kA/m and a $(BH)_{max}$ of about 181.4kJ/m^3 at 200 ℃ as shown in Table 1. The temperature coefficient (β) of sample *B* is about − 0.18%/ ℃. According to Eqn. (2), the maximum operating temperature of sample *B* is about 478 ℃. Since we don't have the capabilities to measure magnetic properties at temperature over 200 ℃, we had to rely on the irreversible loss measurements. The curve of irreversible loss vs temperature of sample *B* is shown in Fig. 1. The irreversible loss of sample *B* increases greatly at about 500 ℃. This value agrees well with the *MOT* value, which is greatly higher than the *MOT* (300 ℃) of commercial Sm_2Co_{17} alloys.

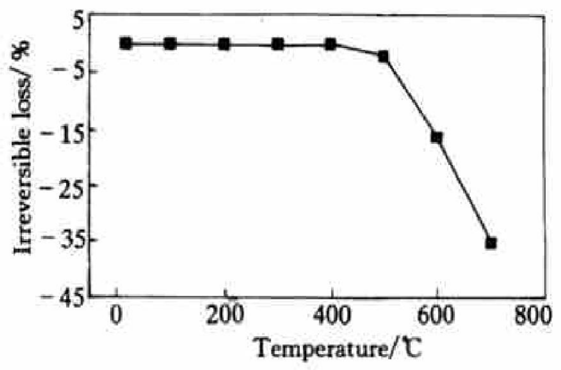

Fig. 1 Irreversible loss of magnet *B* as a function of temperature

3.2 Microstructure analysis

The X-ray patterns of $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnets after fully processed are shown in Fig. 2. A mainly rhombohedral Th_2Zn_{17} phase, $Sm_2(Co, Fe)_{17}$, is identified. Hexagonal $Sm(Co, Cu)_5$ and Co_2Zr are also found in the three magnets.

Fig. 3 shows the SEM images of the $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnets. The microstructure of the magnets consists of cells and cell walls.

The chemical composition of the cells and cell walls determined by EDAX of the 2∶17 magnets is

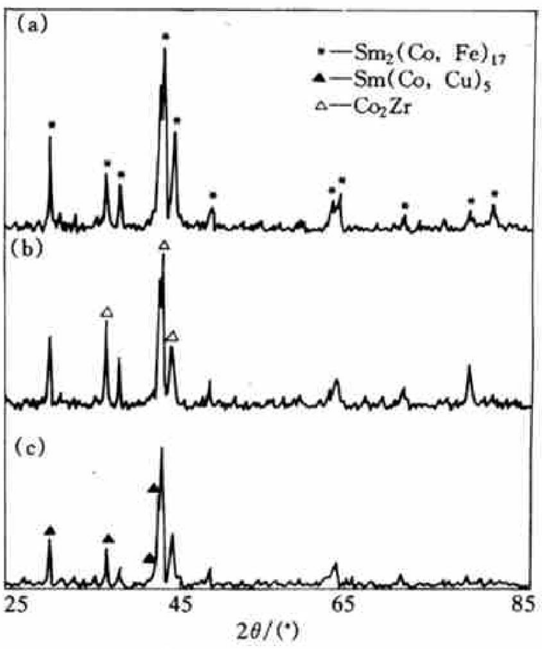

Fig. 2 XRD patterns of $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnets
(a) —Sample *A*; (b) —Sample *B*; (c) —Sample *C*

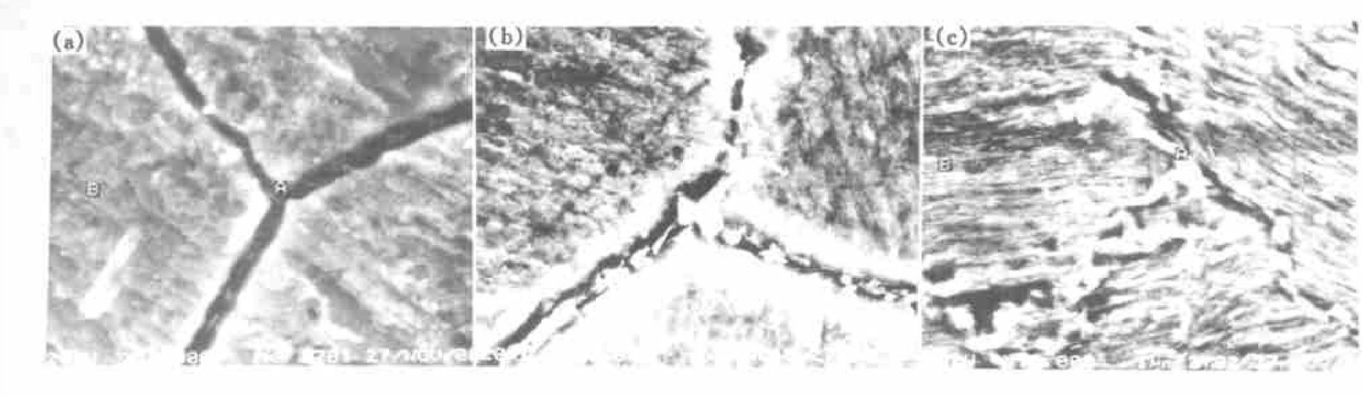

Fig. 3 SEM images of three samples
(a) —Sample *A*; (b) —Sample *B*; (c) —Sample *C*

shown in Table 2. In sample *B*, the cells are enriched in Fe, but depleted in Cu and Zr, whereas the cell boundaries are Cu and Zr enriched, but Fe depleted. This fits well with other studies[5]. There is little difference between samples *A* and *B*. But, in sample *C*, the content of Cu is higher and that of Fe is a little lower in cells.

Table 2 Chemical compositions of cells and cell walls determined by EDAX of $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnets (mole fraction, %)

Sample	Cell walls				
	Sm	Co	Fe	Cu	Zr
A	7.80	65.20	14.70	9.51	2.78
B	8.08	55.96	14.56	13.89	7.51
C	11.33	56.45	22.75	5.19	4.27

Sample	Cells				
	Sm	Co	Fe	Cu	Zr
A	8.85	66.86	15.11	7.37	1.82
B	11.44	55.69	23.53	6.94	2.40
C	13.06	53.52	20.60	10.64	2.14

The coercivity and temperature coefficient of Sm_2Co_{17} magnets are supposed to be related to the microstructure and microchemistry. $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnets are well established as pinning type magnets[6, 7]. Their final microstructure consists of a mixture of cellular and lamella structures[8]. The cell interior is a rhombohedral $Sm_2(Co, Fe)_{17}$ phase surrounded by a hexagonal $Sm(Co, Cu)_5$ cell boundary phase, which pins the domain walls. The Zr-rich lamella phase, for example Co_2Zr, can provide diffusion paths for Cu and help to form the Cu-rich $Sm(Co, Cu)_5$ cell boundary phase. It can lead to a fine uniform celluar microstructure and thus improves its coercivity and temperature dependence[9, 10].

Fig. 4 shows the microstructures of sample *B*. There are two different phases in the magnet, one is black phase, another is white phase. Further analyses show that the white phase is of 2:17 phase precipitate of Fe-rich and Co-rich while the black is of Cu-rich phase precipitate. We also can find a small amount of lamella phase, which is full of Zr.

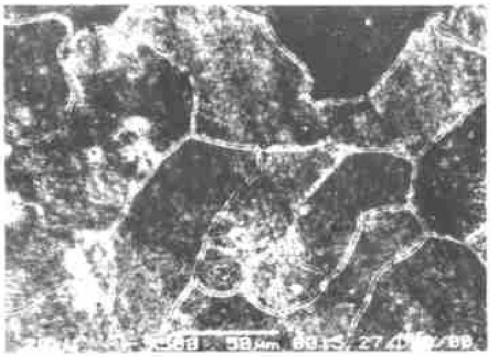

Fig. 4 SEM image of $Sm_2(Co, Cu, Fe, Zr)_{17}$ magnets *B* after fully processed

4 CONCLUSION

We have designed a high-performanced $Sm_2(Co, Fe, Cu, Zr)_{17}$ magnet. The magnetic properties at room temperature are as such, $B_r = 1.08\,T$, $H_{ci} = 2312.0\,kA/m$, $(BH)_{max} = 211.0\,kJ/m^3$. High temperature magnetic measurements reveal that it can be used up to 450 ℃. The microstructure analysis shows that the magnet consists of $Sm_2(Co, Fe)_{17}$, $Sm(Co, Cu)_5$ and Co_2Zr phases. The cells are enriched in Fe, but depleted in Cu and Zr, whereas the cell boundaries are Cu and Zr enriched, but Fe depleted.

[REFERENCES]

[1] Andrew S K. Design of high temperature permanent magnets [J]. J Appl Phys, 1997, 81(8): 5609-5611.

[2] Liu J F, Zhang Y, Ding Y, et al. Rare earth permanent magnets for high temperature applications [A]. Proc 15th Int Workshop on REPM [C]. 1998, 607-622.

[3] Liu J F, Zhang Y, Hadjipanayis G C. High-temperature magnetic properties and microstructural analysis analysis of Sm(Co, Fe, Cu, Zr)z permanent magnets [J]. Journal of Magnetism and Magnetic Materials, 1999, 202(4): 69-76.

[4] Tang W, Zhang Y, Hadjipanayis G C. Microstructure and magnetic properties of $Sm(Co_{bal}Fe_xCu_{0.128}Zr_{0.02})_{7.0}$ magnets with Fe substitution [J]. Journal of Magnetism and Magnetic Materials, 2000, 221(3): 268-272.

[5] Rabenberg L, Mishra R K, Thomas G. Microstructures of precipitation-hardened SmCo permanent magnets [J]. J Appl Phys, 1982, 53(3): 2389-2391.

[6] SUN Tian-duo. A modle on the coercivity of the hardened 2:17 rare earth-cobalt permanent magnets [J]. J Appl Phys, 1981, 52(3): 2532-2534.

[7] Chui S T. Towards a more analytic understanding of the pinning of domain walls in high-temperature permanent magnets [J]. Journal of Magnetism and Magnetic Materials, 2000, 217(2): 120-128.

[8] Kumar K. $RETM_5$ and RE_2TM_{17} permanent magnets development [J]. J Appl Phys, 1988, 63(6): 13-57.

[9] Tang W, Zhang Y, Hadjipanayis G C. High-temperature magnetic properties of $Sm(Co_{bal}Fe_{0.1}Cu_{0.088}Zr_x)_{8.5}$ magnets [J]. J Mag Mag Mater, 2000, 212(1): 138-144.

[10] Tang W, Zhang Y, Goll D. New $Sm(Co, Fe, Cu, Zr)_z$ magnets with better temperature stability [J]. Journal of Magnetism and Magnetic Materials, 2001, 226(3): 1365-1366.

(Edited by HE Xue-feng)