Article ID: 1003－6326(2004)04－0790－04

Microstructure of Sm_2Co_{17} magnets and its influence on coercivity①

LI Li-ya(李丽娅), YI Jian-hong(易健宏), HUANG Bai-yun(黄伯云),
PENG Yuan-dong(彭元东), DU Juan(杜 娟)
(State Key Laboratory of Powder Metallurgy, Central South University, Changsha 410083, China)

Abstract: $Sm(Co_{bal}Fe_{0.197}Cu_{0.049}Zr_{0.026})_{7.5}$ magnet with H_{ci} of 2105 kA/m and $\beta_{18-200\ ℃}$ of －0.17%/℃ was made by sintering processing. The magnet has uniform celluler structure. The cell interior is a rhombohedarl 2∶17 phase, and the boundary is a hexagonal 1∶5 phase. The average cell size is 93 nm and the cell boundary thickness is 20 nm. The cells are enriched in Fe, and the cell boundaries are enriched in Cu. More Cu-riched 1∶5 cell boundary phase would be helpful to obtain a higher coercivity and lower temperature coefficient. White secondary phase mainly consisting of Sm can decrease the coercivity of the magnets, but the closely paralleled grooves can increase coercivity.

Key words: high temperature magnet; coercivity; cell structure; grooves
CLC number: TM 273 **Document code:** A

1 INTRODUCTION

Permanent magnets capable of operating at high temperatures(⩾400 ℃) are required for advanced power systems[1]. Since they combine the highest Curie temperature and crystalline anisotropy, permanent magnet materials based on Sm_2Co_{17} compound are the best candidates among all known materials for the required high temperature applications[2, 3].

The microstructures of Sm_2Co_{17} magnets consist of two phase cellular morphology where the 2∶17 phase forms the cell interiors and the 1∶5 phase the cell boundaries[4, 5]. It was reported that precipitations inside the 2∶17 main phase cells pin the domain wall movement and thus enhance the coercivity[6]. It was also reported that the 1∶5 cell boundaries impede domain wall movement which has a similar effect to that of homogeneous wall pinning[7, 8]. The low H_{ci} magnets generally exhibit smaller cell structure and the high H_{ci} magnets exhibit larger cell structure[9]. However, it has not been understood very well in the studies that the effect of the secondary phase, phase boundary and other crystal defections on coercivity[10]. In this paper, we have investigated the properties of Sm_2Co_{17} magnets and argued the factors that influence the coercivity through TEM, SEM and other analysis methods.

2 EXPERIMENTAL

Samples with nominal composition $Sm(Co_{bal}Fe_xCu_yZr_z)_{7.5}$ with $x=0.197-0.282$, $y=0.049-0.074$ and $z=0.024-0.026$ were prepared by arc-melting. Alloy ingots were ball milled and then compacted in a magnetic field. The green compacts were vaccum sintered at 1 180 to 1 210 ℃ for about 60－120 min and solution treated for 70－120 min at 1 160－1 190 ℃. The further precipitation hardened at 800－860 ℃ for 9－20 h and slowly cooled to 350－400 ℃ at a cooling rate of 0.5－1.5 ℃/min. Magnetic properties were measured using a vibrating sample magnetometer with a maximum applied field of 2 387 kA/m. Microstructure analysis was carried out using transmission electron microscope(TEM) with high resolution energy-dispersive X-ray spectrometer (EDX), scanning electron microscopy(SEM), X-ray diffractometer(XRD), respectively.

3 RESULTS AND DISCUSSION

Table 1 shows the intrinsic coercivity at room temperature and 200 ℃ and the temperature coefficent of the magnets. It can be seen that samples A, B and C, which have higher Cu and Zr contents but lower Fe content, have much higher intrinsic coercivity and lower temperature coefficient. Coercivity and temperature coefficient are decreased with increasing Fe content. But as Cu content increases, the intrinsic coercivity is also lightly increased. According to Tang et al[11], the addition of Zr leads to a fine uniform cellular microstructure and thus improves coercivity. The addition of Fe leads to the disappearance of the cellular microstructure and deterioration of coercivity[12]. Increasing Cu content leads to a higher domain wall energy gradient along the cell boundaries and thus to a higher coercivity[13].

Sm_2Co_{17} type magnet is domain wall pinning type magnet. At the heat demaganitization state, domain wall is pined by pinning dots. The pinning cen-

① **Foundation item:** Project(02JJY2079) supported by the Natural Science Foundation of Hunan Province
Received date: 2003－12－16; **Accepted date:** 2004－04－14
Correspondence: YI Jian-hong, Professor, PhD; Tel: +86-731-8877328; E-mail: pmmagnet@mail.csu.edu.cn

ter could be second phase, phase boundary and other crystal defects, such as crystal boundary, vacancy. It is well established that the coercivity of Sm_2Co_{17} magnets is due to a pinning at the Cu-rich 1∶5 cell boundaries. The cell size, cell form and the thickness of cell boundary control the coercivity of Sm_2Co_{17} magnets. Fig. 1 shows the TEM image and electron diffraction patterns of sample A, respectively. We can draw that sample A has a uniform cellular microstructure. The average cell size is 93 nm and cell boundary thickness is 20 nm. From the electron diffraction pattern, we can draw that the cellular structure consists of 2∶17*H* phase, 2∶17*R* phase and 1∶5 phase. The cell interior is a rhombohedarl 2∶17 phase, and the boundary is a hexagonal 1∶5 phase. The 2∶17*R* phase is absolutely separated by the 1∶5 phase. Fig. 2 shows the EDX profiles of the elements Cu and Fe within the 1∶5 cell wall and the 2∶17*R* cell, respectively. From the analysis of the element, the cells are enriched in Fe, and the cell boundaries are enriched in Cu. With more Cu substituted in, the 1∶5 cell boundary phase becomes close to nonmagnetic, resulting in complete magnetic isolation of the 2∶17*R* cells and thus in a high room temperature coercivity[14].

Fig. 3 shows the X-ray patterns of samples A, B, C, D and E. Similar to the results of electron diffraction pattern, a mainly rhombohedral 2∶17 phase and a hexagonal 1∶5 phase are found in those alloys, but the diffraction intensity of 2∶17*R* phase and 1∶5 phase are different from these samples. We can see that, samples A and B have the highest diffraction intensity of 1∶5 phase, and the diffraction intensity of 1∶5 phase of sample E is the lowest. The $CaCu_5$-type Cu-rich 1∶5 cell boundary phase is responsible for the high intrinsic coercivity by pinning the domain

Table 1 Intrinsic coercivity and temperature coefficient β

Sample	Composition	H_{ci}/(kA·m^{-1})		$\beta_{18\text{-}200\ ℃}$/(%·℃$^{-1}$)
		20 ℃	200 ℃	
A	$Sm(Co_{bal}Fe_{0.197}Cu_{0.124}Zr_{0.026})_{7.5}$	2 105	1 472	− 0.17
B	$Sm(Co_{bal}Fe_{0.197}Cu_{0.074}Zr_{0.026})_{7.5}$	2 160	1 432	− 0.19
C	$Sm(Co_{bal}Fe_{0.197}Cu_{0.049}Zr_{0.026})_{7.5}$	2 088	1 352	− 0.20
D	$Sm(Co_{bal}Fe_{0.197}Cu_{0.049}Zr_{0.024})_{7.5}$	1 928	1 128	− 0.23
E	$Sm(Co_{bal}Fe_{0.239}Cu_{0.049}Zr_{0.024})_{7.5}$	1 808	984	− 0.25
F	$Sm(Co_{bal}Fe_{0.282}Cu_{0.049}Zr_{0.024})_{7.5}$	1 672	832	− 0.27

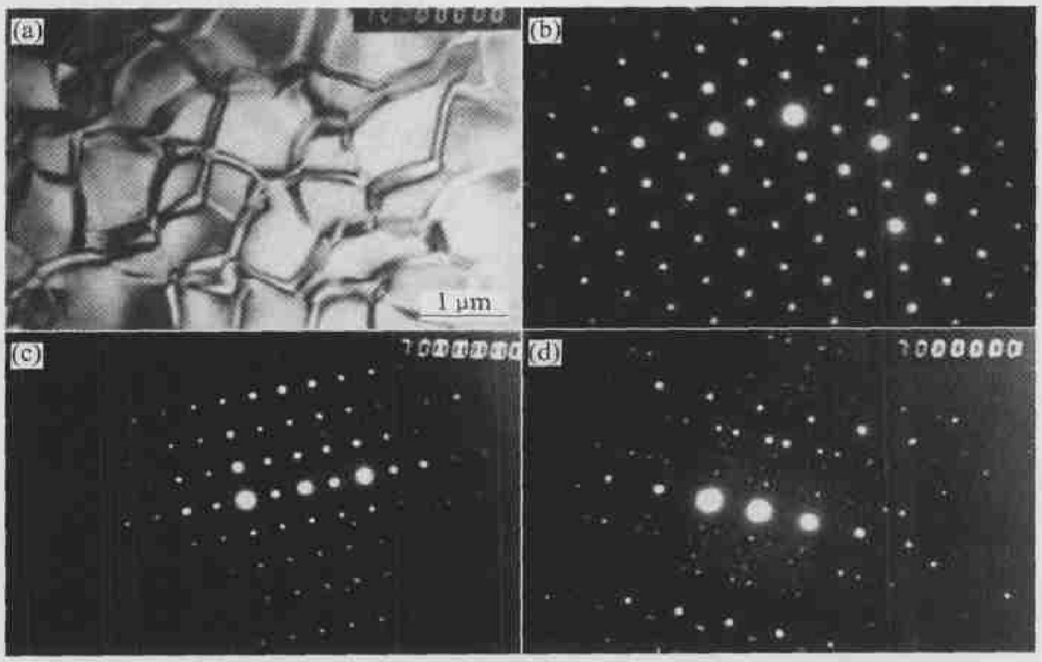

Fig. 1 TEM micrograph and electron diffraction patterns of magnet $Sm(Co_{bal}Fe_{0.197}Cu_{0.124}Zr_{0.026})_{7.5}$
(a) —TEM photograph; (b) —Electron diffraction pattern of 2∶17*H* phase;
(c) —Electron diffraction pattern of 2∶17*R* phase; (d) —Electron diffraction pattern of 2∶17*R* and 1∶5 phase

walls. More 1∶5 phase would be helpful to obtain a higher coercivity. So the coercivities of samples A and B are much higher than that of sample E.

Fig. 4 shows the SEM microstructures of the alloys. It can be drawn that, besides matrix, there are white phase, small holes and grooves in the magnets. The EDX analysis of white phase shows that it contains Sm 94.98%, Co 4.02%, Fe 0.90% and Zr 0.20%. We can find that the white phase is mainly consisted of Sm. Sm precipitated from the matrix because of the high sintering temperature, which makes Sm volatilized from the matrix[15]. The decrease of Sm content in the magnetic phase will reduce the square of demagnetization curve and affect the maximum energy and coercivity. At the other hand, we can find that there are many closely paralleled grooves in the crystals. But the orientation of paralleled grooves is different from crystals. And the same result is found when observed at the single polarized light and colour polarized light through metallurgy micrograph. There are much more paralleled grooves in the crystals of the magnets having high coercivity, such as magnets A and B, but we hardly can find any grooves in magnet E. Those grooves could be looked as crystal defects. When domain wall moves to grooves, they would be pinned. More domain wall pinning sites would be helpful to obtain a higher coercivity.

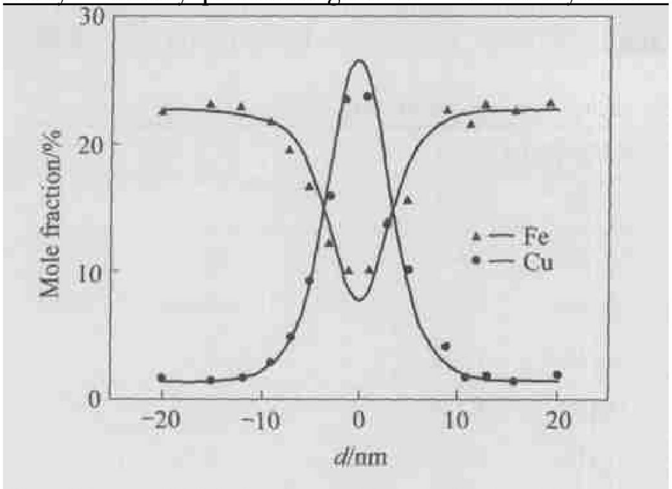

Fig. 2 EDX profiles of elements Cu and Fe within 1∶5 cell wall and 2∶17 cell of magnet $Sm(Co_{bal}Fe_{0.197}Cu_{0.124}Zr_{0.026})_{7.5}$

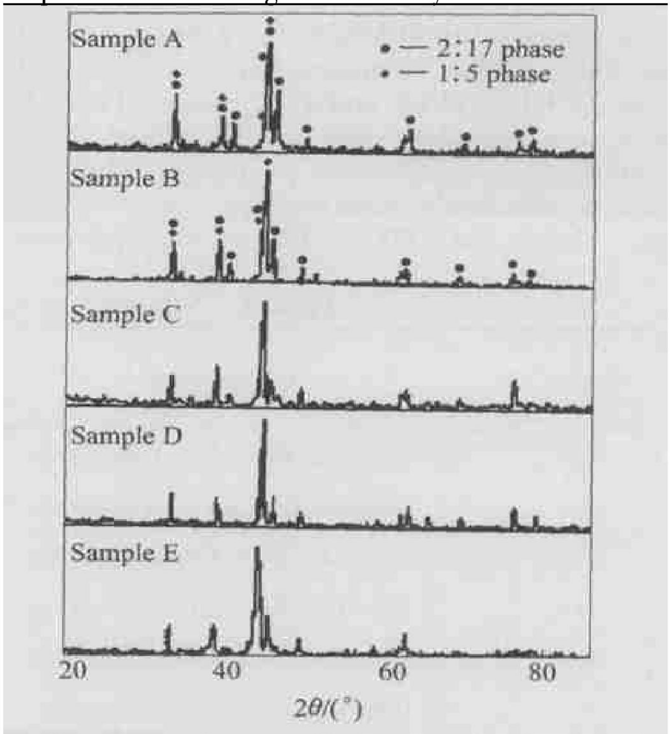

Fig. 3 X-ray diffraction patterns of samples A, B, C, D and E

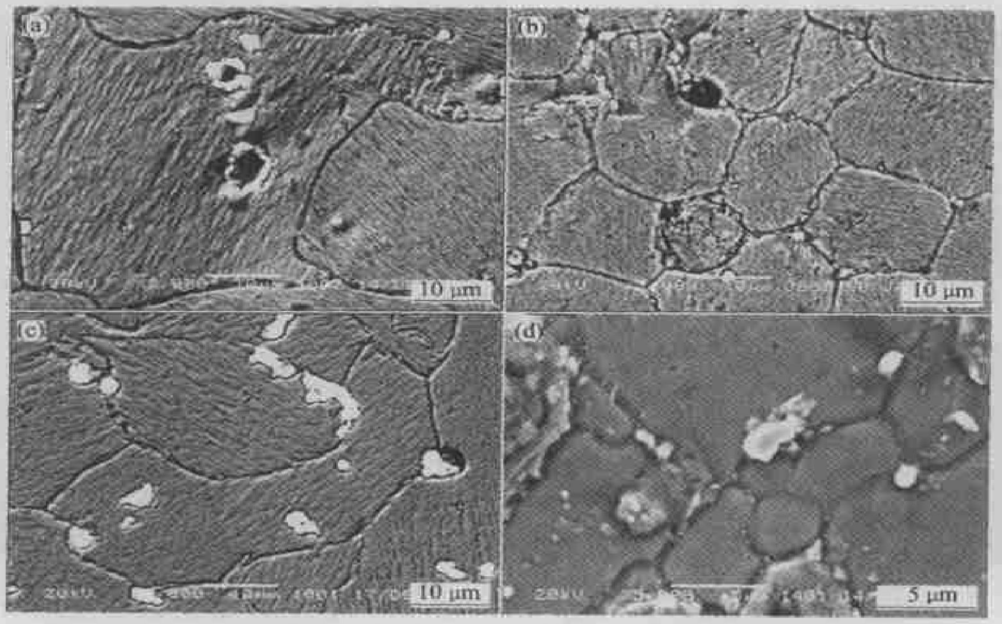

Fig. 4 SEM images of full treated magnets
(a) —Sample A; (b) —Sample B; (c) —Sample C; (d) —Sample E

4 CONCLUSIONS

1) $Sm(Co_{bal}Fe_{0.197}Cu_{0.049}Zr_{0.026})_{7.5}$ magnet with H_{ci} of 2 105 kA/m and $\beta_{18-200\ ℃}$ of − 0.17%/℃ has uniform celluler structure. The cell interior is a rhombohedarl 2∶17 phase, and the boundary is a hexagonal 1∶5 phase. The average cell size is 93 nm and cell boundary thickness is 20 nm. The cells are enriched in Fe, and the cell boundaries are enriched in Cu. More 1∶5 cell boundary phase would be helpful to obtain a higher coercivity and lower temperature coefficient.

2) White secondary phase mainly consisting of Sm decrease the coercivity of the magnets, but the closely paralleled grooves could increase the coercivity.

REFERENCES

[1] WANG Cheng-de. 550 ℃ high temperature permanent magnets[J]. Research in Metallic Material, 2001, 27(2): 55−61. (in Chinese)

[2] Hadjipanayis G C, Tang W, Zhang Y, et al. High temperature 2∶17 magnets: relationship of magnetic properties to microstructure and processing[J]. IEEE Trans On Mag, 2000, 36(5): 3382−3387.

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

[4] Matthiasa T, Zehetnera G, Fidler J. TEM-analysis of $Sm(Co, Fe, Cu, Zr)_z$ magnets for high-temperature applications[J]. Journal of Magnetism and Magnetic Materials, 2002, 242−245 (2) : 1353−1355.

[5] Kronmuller H, Goll D. Micromagnetic analysis of pinning-hardened nanostructured, nanocrystalline Sm_2Co_{17} based alloys[J]. Scripta Materialia, 2002, 47(4): 545−550.

[6] Streibl B, Filder J, Schrefl T. Domain wall pinning in high temperature $Sm(Co, Fe, Cu, Zr)_{7-8}$ magnets[J]. Journal of Applied Physics, 2000, 87(9): 4765−4767.

[7] Yong Z, Michelle C R, George C H. Magnetic hardening studies in sintered $Sm(Co, Cu_x, Fe, Zr)_z$ 2∶17 high temperature magnets[J]. Journal of Applied Physics, 2000, 87(9): 6722−6724.

[8] Xiong X Y, Ohkubo T, Koyama T, et al. The microstructure of sintered $Sm(Co_{0.72}Fe_{0.20}Cu_{0.055}Zr_{0.025})_{7.5}$ permanent magnet studied by atom probe[J]. Acta Materialia, 2004, 529(5): 737−748.

[9] Liu J F, Zhang Y, Dimitrov D, et al. Microstructure and high temperature magnetic properties of $Sm(Co, Cu, Fe, Zr)_z$ (z = 6.7−7.1) permanent magnets[J]. Journal of Applied Physics, 1999, 85(5): 2800−2804.

[10] ZHOU Shou-zeng. Rare-earth Permanent Magnets and Their Application[M]. Beijing: Metallurgical Industry Press, 1998. 309−324. (in Chinese)

[11] Tang W, Zhang Y, Hadjipanayis G C. Effect of Zr on the microstructure and magnetic properties of $Sm(Co_{bal}Fe_{0.1}Cu_{0.088}Zr_x)_{8.5}$ magnets[J]. J Appl Phy, 2000, 87(2): 399−403.

[12] 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(1): 268−272.

[13] Zhang Y, Tang W, Hadjipanayis G C, et al. Evolution of microstructure, microchemistry and coercivity in 2∶17 type Sm-Co magnets with heat treatment[J]. IEEE Transactions on Magnetics, 2001 , 37 (4): 2525−2527.

[14] Tang W, Zhang Y, Gabay A M, et al. Anomalous temperature dependence of coercivity in rare earth cobalt magnets[J]. Journal of Magnetism and Magnetic Materials, 2002, 242−245(1): 1335−1337.

[15] LI Li-ya, YI Jian-hong, ZENG Qing-ling. Investigation of high temperature rare earth permanent magnets $Sm_2(Co, Cu, Fe, Zr)_{17}$[J]. Chinese Journal of Materials Research, 2003, 17(1): 45−49. (in Chinese)

(Edited by LONG Huai-zhong)