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ARTICLE

Influence of Fe Content on Magnetic Properties of High Temperature Rare Earth Permanent Magnets $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$($x$=0.09-0.21)

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Abstract: The influence of Fe content on magnetic properties of high temperature rare earth permanent magnets $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09-0.21), prepared by the conventional powder metallurgy method, has been studied. The results show that: with increasing of Fe content, B_r and $(BH)_{max}$ increased, whose optimum value reached to 0.96 T and 176.7 kJ/m^3 while x=0.21; H_{ci} increased first, but then decreased with further increasing of Fe content; the optimum value of H_{ci} reached to 2276.6 kA/m while x=0.15. The temperature stability of the magnet with x=0.15, prepared by optimum process, has been proved to be excellent, and the B-H demagnetization curve kept a straight line at 500 °C; the intrinsic coercivity coefficient β was –0.16%/°C, and the maximum operating temperature was up to 533 °C.

Key words: high temperature rare earth permanent magnet; $Sm(Co,Fe,Cu,Zr)_z$ magnet; magnetic property; temperature stability

In recent years, there has been a demand for permanent magnets which can be used at up to ≥ 400 °C [1-3]. $Sm(Co, Fe, Cu, Zr)_z$ permanent magnets, combining the highest Curie temperature with moderately high saturation magnetization and crystallization anisotropy, are the most promising candidates for high temperature applications[4-6]. But the maximum operating temperature of commercial $Sm(Co,Fe,Cu,Zr)_z$ permanent magnet is about 300 °C. With increasing of temperature, the coercivity of commercial $Sm(Co,Fe,Cu,Zr)_z$ permanent magnets decreases rapidly when temperature is above 300 °C, which limits the applications of these magnets. So, the main intention is to increase the maximum operating temperature.

The maximum operating temperature$(OT)_{max}$ of $Sm(Co,Fe,Cu,Zr)_z$ permanent magnets is calculated with formula (1)

$$(OT)_{\max} = RT - \frac{H_{\rm ci,RT} - (H_{\rm ci,OT})_{\min}}{H_{\rm ci,RT} \times \beta} \tag{1}$$

$$\beta = \frac{H_{\rm ci,RT} - H_{\rm ci,OT}}{H_{\rm ci,RT} \times (RT - OT)} \times 100\ \% \tag{2}$$

where RT is room temperature, $H_{ci,RT}$ and $H_{ci,OT}$ denote intrinsic coercivity at room temperature and maximum operating temperature respectively, and β is the temperature coefficient of intrinsic coercivity between RT and OT. It is clear to see from formula (1) that there are two ways to increase $(OT)_{max}$ e. g. increasing $H_{ci,RT}$ or decreasing β and the later is more effective. It is safe to set $(H_{ci,OT})_{min}$=400 kA/m for high temperature applications[6].

Previous studies[1,6-8] showed that the Fe content has great effects on the magnetic properties and temperature stability. The aim of the present paper is to study the influence of Fe content of the magnetic properties. $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09~0.21) magnets were prepared by vacuum induction melting and conventional powder metallurgy processing.

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1 Experimental

Using pure (>99.5%) Sm, Co, Fe, Cu, and Zr alloys as starting materials, the permanent magnets with nominal compositions of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09-0.21) were prepared. The components of the magnets are shown in Table 1. The procedure of sample preparation was as follows: (1) the preparation of magnetic powder, (2) the preparation of sintered magnet samples, and (3) heat treatment. The alloy cast ingots were obtained by melting process in Ar atmosphere in an induction furnace, and then the ingots were crushed to get the powder with average particle size of 4-6 μm. The powder was pressed into compacts in a magnetic field of 800 kA/m; afterwards, the compacts were pressed further by cold isostatic compression at 200 MPa, presintered at 1200 °C for 30 min in vacuum, and sintered into the magnet samples at 1205-1225 °C for 45 min in Ar atmosphere. The samples were solution-heat-treated at 1185 °C for 60 min, and subsequently annealed at 810 °C for 10 h followed by slow cooling at 0.7 °C /min to 400 °C, then in air cooling.

The samples were machined into the cylindric samples with ϕ100 mm(diameter)×10 mm (length). The room temperature and high temperature magnetic properties of ϕ10 mm×10 mm cylindric samples were measured using NIM-10000 hysteresigraph and improved NIM-2000 hysteresigraph, respectively. In order to study irreversible flux Loss, some the samples were machined into the cylindric sample with ϕ10 mm×7 mm, which were aged at 25-650 °C for 2 h in air. The flux-difference was measured by DGY-2B permanent magnetic tester. The element contents were analyzed by EDA×9100160 type energy dispersive analyzer.

2 Results and Discussion

For $Sm(Co,Fe,Cu,Zr)_z$ permanent magnets, the optimum sintering temperature varies with the Fe content. The optimum magnetic properties of the $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09-0.21) magnets are listed in Table 2. It can be seen that the B_r and $(BH)_{max}$ increase with increasing of Fe content, and the maximum value of 0.96 T and 176.7 kJ/m^3 are obtained at x=0.21. When x increases from 0.09 to 0.15, the B_r and $(BH)_{max}$ increase swiftly. But the B_r and $(BH)_{max}$ have a slight increase with x further increasing after from 0.09 to 0.15. In $Sm_2(Co_{1-x}Fe_x)_{17}$ structure, Co atoms can be substituted by Fe atoms. With increasing of Fe content, the saturation magnetization B_s of $Sm_2(Co_{1-x}Fe_x)_{17}$ phase increases because of the higher atomic magnetic moment of Fe element. This is regarded as the main reason for higher B_r and $(BH)_{max}$ with more Fe content. It is also clear that the H_{ci} increases first and then decreases with further increasing of Fe content, and the maximum value of 2276.6 kA/m can be found at x=0.15. Many factors affect the H_{ci} of $Sm(Co,Fe,Cu,Zr)_z$ magnets, as the detailed discussion shown in reference[7].

Table 1 Components of the magnets (ω/%)

Fe content, x	Sm	Co	Fe	Cu	Zr
0.09	25.02	57.36	6.27	7.93	3.42
0.15	25.08	53.07	10.48	7.95	3.42
0.21	25.14	48.76	14.70	7.97	3.43

Table 2 Optimum magnetic properties of magnets

Fe content, x	t/°C	B_r/T	H_{ci}/kA·m^{-1}	$(BH)_{max}$/kJ·m^{-3}
0.09	1225	0.84	2109.4	131.3
0.12	1225	0.90	2228.8	152.0
0.15	1215	0.94	2276.6	171.9
0.18	1210	0.95	2149.2	173.4
0.21	1210	0.96	1948.4	176.7

The Sm content has significant effects on the H_{ci} of $Sm(Co,Fe,Cu,Zr)_z$ magnets. The Sm contents of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$($x$=0.09-0.21) magnets prepared by optimal process are listed in Table 3. It is clear that the Sm content of the magnets with x=0.09 is obviously lower than that of the magnets with x=0.15 and 0.21. This is because the sintering temperature for the magnets with x=0.09 is high up to 1225 °C. During the sintering process, the Sm element will vaporize for its lower melting point even in the atmosphere of Ar gas. According to the phase diagram[8], the amount of 1:5 phase $[Sm(Co_{1-x}Cu_x)_5]$ would decrease with decreasing of Sm content, resulting in decreasing of H_{ci}.

The H_{ci} is correlated with the $\Delta\gamma$ of $Sm_2(Co_{1-x}Fe_x)_{17}$ phase and $Sm(Co_{1-x}Cu_x)_5$ phase. High $\Delta\gamma$ leads to high H_{ci}[9]. The γ is determined by K_1 and A[10]:

$$\gamma = 4\sqrt{K_1 \times A} \tag{3}$$

The $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09-0.21) magnets have a high Cu content (≈8wt%). The pinning structure at RT belongs to attracting type, so the $\Delta\gamma=\gamma^{2:17}-\gamma^{1:5}$ ($\Delta\gamma>0$). The $K_1^{2:17}$ and $A^{2:17}$ would decrease with increasing of Fe content, leading to the decrease of $\gamma^{2:17}$ and $\Delta\gamma$, so does the H_{ci}.

Therefore, the sintering temperature decreases with increasing of Fe content, which is beneficial to the decrease of Sm vaporization, leading to high H_{ci}, when x varies from 0.09 to 0.15. When x vary from 0.15 to 0.21, the Sm content loss is nearly the same for the lower sintering temperature, but the $\Delta\gamma$ decreases with increasing of Fe content, leading to low H_{ci}.

2.2 High temperature magnetic properties and temperature stability

Fig.1 shows the high temperature demagnetization curves of the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets. The B-H demag-

Table 3 Sm content of magnets

Fe content, x	t/°C	Sm content, ω/%
0.09	1225	22.35
0.15	1215	26.34
0.21	1210	26.37

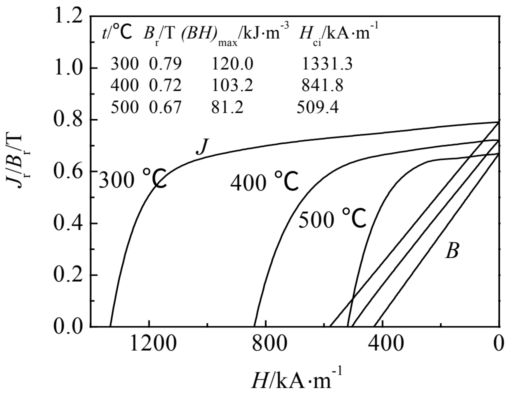

Fig.1 High temperature demagnetization curves of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets

netization curve nearly keeps straight at 500 °C, exhibiting good temperature stability. According to formula (1) and (2), the β and $(OT)_{max}$ of $Sm(Co_{0.72}Fe_{0.15}$- $Cu_{0.1}Zr_{0.03})_{7.5}$ magnets are calculated to be –0.16 %/℃ and 533 ℃, respectively, while the β and $(OT)_{max}$ of commercial $Sm(Co,Fe,Cu,Zr)_z$ magnets are –0.30%/℃ and 533 °C, respectively.

The high temperature magnetic properties of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets and commercial $Sm(Co, Fe, Cu, Zr)_z$ magnets are compared in Fig.2. It can be seen that the effects of temperature on the B_r is similar for the two types of magnets. The Fe content of the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets is lower than that of the commercial $Sm(Co,Fe,Cu,Zr)_z$ magnets, so the lower B_r is obtained for the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets. The lower $(BH)_{max}$ is observed when temperature varies from 25 to 400 °C; only when the temperature arrives to 500 °C, the $(BH)_{max}$ is higher than that of the commercial $Sm(Co,Fe,Cu,Zr)_z$ magnets. These two types of magnets have similar H_{ci} at room temperature, but the H_{ci} of the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets decreases move slowly than that of the commercial $Sm(Co,Fe,Cu,Zr)_z$ magnets; when the temperature arrives to 500 °C, the higher H_{ci} is obtained for the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets.

The temperature stability of magnets defined as the dependence of magnetic properties on temperature. It can be expressed indirectly by the irreversible flux loss η_{Irr}[11,12]:

$$\eta_{Irr} = \frac{\phi' - \phi}{\phi} \times 100\ \% \tag{4}$$

where, ϕ is the flux at room temperature, ϕ' is the flux when samples are exposed at high temperature and measured at room temperature, and commonly the $\phi' < \phi$. So, the $\eta_{Irr} <$ 0 and the absolute value of η_{Irr} is irreversible flux loss.

The η_{Irr} of the $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnet and 2:17 type SmCo magnet prepared by Southwest Institute of Applied Magnetics is shown in Fig.3. The η_{Irr} of the 2:17 type SmCo magnet increases obviously when the temperature arrives to 400 °C, while that is 500 °C for the $Sm(Co_{0.72}Fe_{0.15}$ $Cu_{0.1}Zr_{0.03})_{7.5}$ magnet. The η_{Irr} of the $Sm(Co_{0.72}Fe_{0.15}$ $Cu_{0.1}$ $Zr_{0.03})_{7.5}$ magnet at 550 °C is 3.78% (<5%), exhibiting good

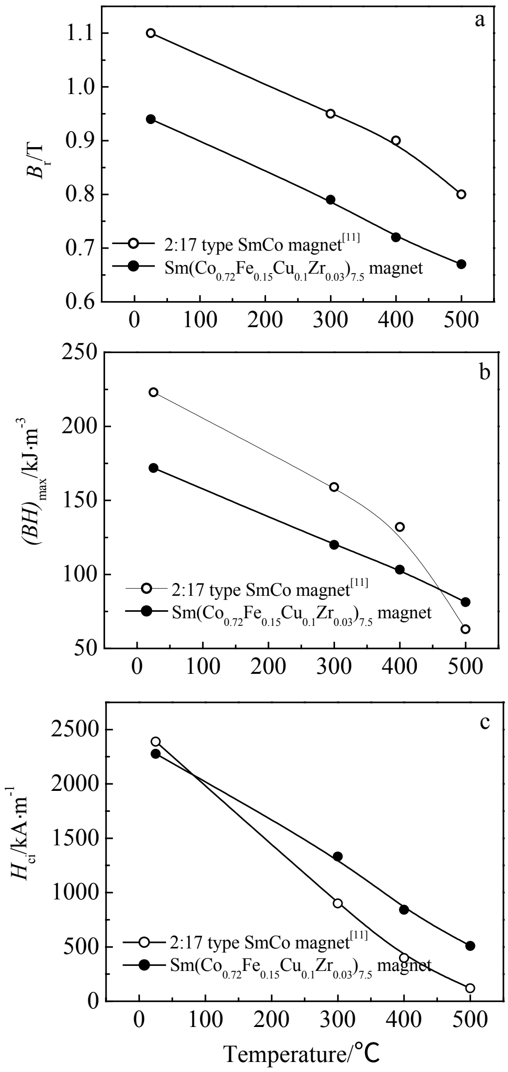

Fig.2 Influence of temperature on magnetic properties of magnets: (a) Br vs temperature, (b) (BH)max vs temperature, and (c) Hci vs temperature

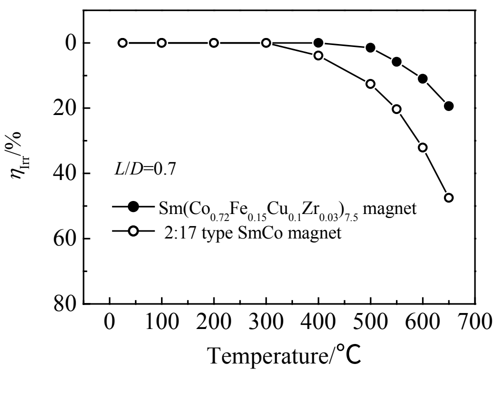

Fig.3 Influence of temperature on irreversible flux loss of magnets

temperature stability, which can be used in high temperature (<550 °C) case.

3 Conclusions

1) The sintering temperature decreases with increasing of Fe content; the sintering temperature is 1225 °C when x<0.15, and the Sm vaporizes seriously, leading to the low magnetic properties.

2) The B_r and $(BH)_{max}$ increase with increasing of Fe content, and the maximum value of 0.96 T and 176.7 kJ/m^3 are obtained at x=0.21. The H_{ci} increases first and then decreases with further increasing of Fe content, and the maximum value of 2276.6 kA/m is observed at x=0.15.

3) The B_r, $(BH)_{max}$ and H_{ci} at 25 °C and 500 °C for $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets prepared by optimum process are 0.94 T, 171.9 kJ/m^3, 2276.6 kA/m and 0.67 T, 81.2 kJ/m^3, 509.4 kA/m, respectively. The magnets exhibits good temperature stability, the *B-H* demagnetization curve nearly keeps straight at 500 °C, and the β and $(OT)_{max}$ are –0.16 %/°C and 533 °C, respectively.

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