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Rare earth permanent magnets $Sm_2(Co, Fe, Cu, Zr)_{17}$ for high temperature applications

PENG Long (彭 龙)[1], YANG Qiuhui (杨青慧)[1], ZHANG Huaiwu (张怀武)[1], XU Guangliang (徐光亮)[2], ZHANG Ming (张 明)[3], WANG Jingdong (王敬东)[3]

(1. State Key Laboratory of Electronic Thin Films and Integrated Devices, University of Electronic Science and Technology of China, Chengdu 610054,China；2. Shcool of Materials Science and Engineering, Southwest University of Science and Technology, Mianyang 621010, China；3. Southwest Institute of Applied Magnetics, Mianyang 621000, China)

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Abstract: Sintered $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09–0.21) permanent magnets with higher Fe content were found to have higher remanence B_r and maximum energy product $(BH)_{max}$ at room temperature. B_r and $(BH)_{max}$ reached maximum of 0.96 T and 176.7 kJ/m^3, respectively at room temperature when the Fe content x reached 0.21. However, the intrinsic coercivity H_{ci} at room temperature increased gradually when the Fe content x increased from 0.09 to 0.15, but when x further increased to 0.21, H_{ci} decreased. H_{ci} attained its peak value of 2276.6 kA/m with Fe content x=0.15 at room temperature. For magnets with x=0.15, B_r, $(BH)_{max}$ and H_{ci} reached 0.67 T, 81.2 kJ/m^3 and 509.4 kA/m at 500 °C, respectively, showing good high temperature stability, which could be used in high temperature applications.

Keywords: rare earth permanent magnets; SmCo alloys; coercivity; temperature coefficient

In recent years, there has been a demand for permanent magnets which can be used at up to ≥450 °C[1–3]. $Sm(Co, Fe, Cu, Zr)_z$ 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]. With rising temperature, the intrinsic coercivity of commercial $Sm(Co, Fe, Cu, Zr)_z$ magnets decreases rapidly when temperature is above 300 °C, it drops to lower than 398 kA/m at 450 °C. Therefore, the magnetic properties of commercial $Sm(Co, Fe, Cu, Zr)_z$ magnets need to be improved for high temperature applications.

Previous studies[1,6–8] showed that high temperature magnetic properties and good temperature stability of $Sm(Co, Fe, Cu, Zr)_z$ magnets can be improved by applying higher Sm content, lower Fe content and higher Cu content in the magnetic alloys. In a previous study, the influence of mean particle size on magnetic properties of sintered $Sm(Co, Fe, Cu, Zr)_z$ magnets was investigated[9]. In this article, high temperature $Sm(Co, Fe, Cu, Zr)_z$ magnets were prepared by adjusting the Fe content and optimizing the processing properly.

1 Experimental

New permanent magnets with compositions of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09–0.21) were prepared by vacuum induction melting and conventional powder metallurgy processing. The room temperature and high temperature magnetic properties were measured using NIM-10000 hysteresigraph and NIM-2000 hysteresigraph, respectively. The irreversible flux loss was estimated by measuring the flux-difference with DGY-2B permanent magnetic tester before and after exposing the samples to elevated temperature for 2 h in air. The element content was analyzed by EDAX9100/60 type energy dispersive analysis.

2 Results and discussion

2.1 Effects of Fe content on magnetic properties

It is found that the optimal sintering temperature is correlated with Fe content for $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09–0.21) magnets. To optimize Fe content and obtain high magnetic properties, the sintering temperature is fixed as optimal value for each different Fe content x. Fig.1 shows the dependence of magnetic properties on Fe content of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09–0.21) magnets. It can be seen that the remanence B_r and maximum energy product

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Corresponding author: ZHANG Huaiwu (**E-mail:** hwzhang@uestc.edu.cn; **Tel.:** +86-28-83207063)

$(BH)_{max}$ increase rapidly when Fe content x increases from 0.09 to 0.15. With x further increasing to 0.21, B_r and $(BH)_{max}$ increase slightly, reaching the maximum value of 0.96 T and 176.7 kJ/m^3 respectively. However, the intrinsic coercivity H_{ci} increases first and then decreases with increasing Fe content, with the peak value of 2276.6 kA/m at x=0.15. Besides, owing to the fact that B_r and $(BH)_{max}$ reach 0.94 T and 171.9 kJ/m^3 respectively at x=0.15, which are very close to each of their maximum values, so the optimal Fe content x should be 0.15.

In $Sm_2(Co_{1-x}Fe_x)_{17}$ structure, Co atoms can be substituted by Fe atoms. With increasing Fe content, the saturation magnetization B_s of $Sm_2(Co_{1-x}Fe_x)_{17}$ phase increases because of higher atomic magnetic moment of Fe element. This is regarded as the main reason for the higher values of B_r and $(BH)_{max}$ with higher Fe content. But the effects of Fe content on H_{ci} are complex. In principle, increased Fe content weakens the exchange interaction and decreases the magnetic anisotropy, resulting in lower domain wall energy gradient $\Delta\gamma$ and H_{ci}. This is consistent with the experimental results when $x\geq 0.15$, but it is not true when $x<0.15$. When $x<0.15$, the optimal sintering temperature is 1225 °C and the Sm content is lower than 23%, which is less than that of magnets with $x\geq 0.15$ sintered at lower optimal sintering temperature. Lower Sm content leads to the decrease of 1:5 cell boundary phases, resulting in lower domain wall pinning strength and H_{ci}.

2.2 Effects of sintering temperature on magnetic properties

The solution temperature is fixed as 1185 °C to investigate the effects of sintering temperature on magnetic properties of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets. Fig.2 presents the dependence of magnetic properties and densities on the sintering temperature of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets. According to Fig.2(a), B_r has a swift increase when the sintering temperature increases from 1205 to 1210 °C. But when the sintering temperature increases from 1210 to 1220 °C, B_r increases slightly, obtaining the peak value at 1220 °C. With the sintering temperature further increasing to 1225 °C, B_r decreases. When the sintering temperature increases from 1205 to 1215 °C, the density D increases rapidly, but it increases slightly when the sintering temperature increases from 1215 to 1220 °C. With the sintering temperature further increasing to 1225 °C, the density is found to be very difficult to increase.

It is well known that B_r of sintered Sm(Co, Fe, Cu, Zr)$_z$

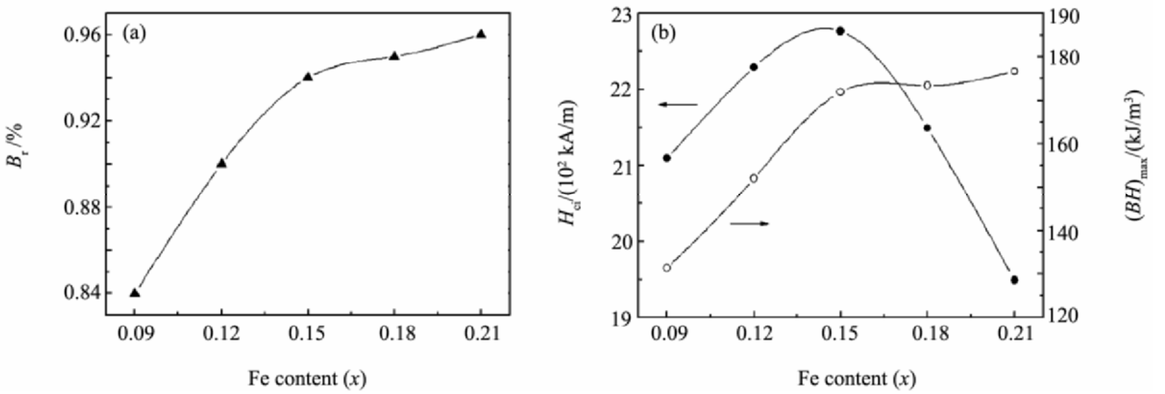


Fig.1 Dependence of magnetic properties on Fe content of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (x=0.09–0.21) magnets
(a) B_r versus Fe content; (b) H_{ci} and $(BH)_{max}$ versus Fe content

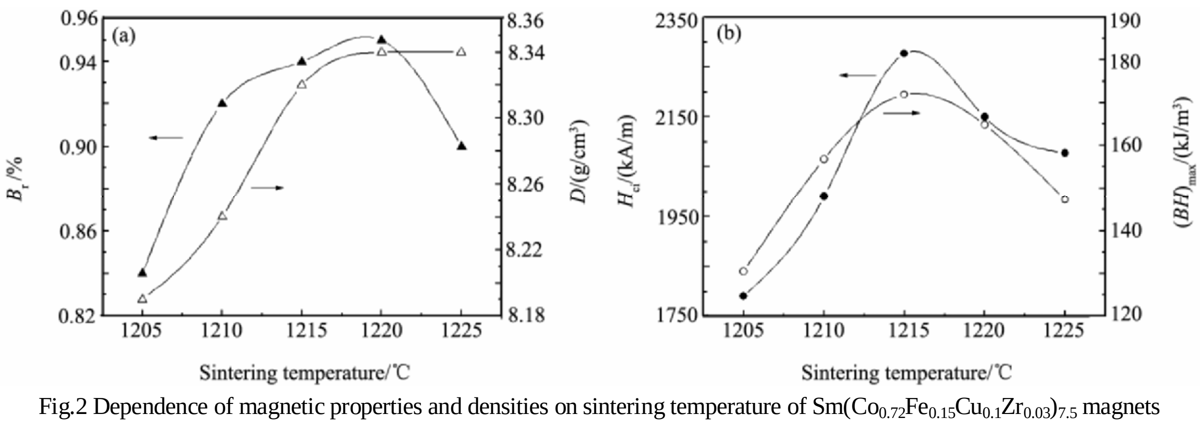


Fig.2 Dependence of magnetic properties and densities on sintering temperature of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets
(a) B_r and D versus sintering temperature; (b) H_{ci} and $(BH)_{max}$ versus sintering temperature

Table 1 Sm content of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets

Sintering temperature/°C	1205	1210	1215	1220	1225
Sm content/%	26.45	26.40	26.34	25.63	22.31

magnets is determined by Eq.(1):

$$B_r = A(1-N)Dm_0M_s\overline{\cos q} \quad (1)$$

where, A, N, D and $\overline{\cos q}$ denote proportion of positive domains, proportion of nonmagnetic phase, density and orientation index, respectively. According to Eq.(1), B_r increases monotonously with D, which can well explain the relationship between Br and D of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets when the sintering temperature increases from 1205 to 1220 °C, but it is invalid when the sintering temperature further increases to 1225 °C. Table 1 shows the Sm content of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets sintered at different temperatures. It is clear that Sm content decreases slightly when the sintering temperature increases from 1205 to 1220 °C, but there is a sharp decrease when the sintering temperature further increases to 1225 °C. According to the Sm-Co phase diagram[10], the amount of 2:17R phase decreases with decreasing Sm content, resulting in lower A and M_s in Eq.(1), which is the main reason for the decrease of B_r when the sintering temperature increases from 1220 to 1225 °C.

In addition, as seen from Fig.2(b), H_{ci} and $(BH)_{max}$ increase first and then decrease with increasing sintering temperature from 1205 to 1225 °C, and their peak values occur at 1215 °C. Therefore, 1215 °C is the optimal sintering temperature for obtaining high magnetic properties of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets.

2.3 Effects of solution temperature on magnetic properties

Fixing the sintering temperature at 1215 °C, the effects of solution temperature on magnetic properties of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets are investigated. Fig.3 shows the dependence of magnetic properties on solution temperature of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets. It can be seen that B_r slightly changes when the solution temperature increases from 1165 to 1195 °C, but when the solution temperature further increases to 1205 °C, B_r decreases sharply. H_{ci} and $(BH)_{max}$ increase first and then decrease with further increase of the solution temperature, their peak values occurs both at 1185 °C. So the optimal solution temperature is found to be 1185 °C.

The purpose of solution is to produce highly supersaturated single phase alloy. The solution temperature has two main effects on coercivity of magnets. On the one side, the relationship between diffusion coefficient D and temperature T can be described as in Eq.(2):

$$D=D_0e^{-Q/RT} \quad (2)$$

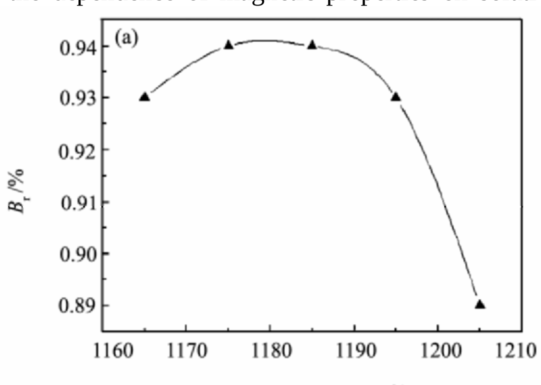
where, D_0 is a constant, Q is activation energy and R is gas constant. With the same solution time, higher T leads to higher D, which is beneficial for the solution of different phases and is easier to obtain a highly supersaturated single phase. The complex cell configuration and small cell size of magnets can be easily formed during isothermal aging if the alloy has a high degree of supersaturation obtained during solution treatment. The relationship between H_{ci} and cell size d can be simply described as in Eq.(3):

$$H_{ci}=\frac{\sqrt{3}\Delta\gamma}{2M_sd} \quad (3)$$

where, $\Delta\gamma$ denotes the domain wall energy gradient between 2:17 cell phase and 1:5 cell boundary phase. According to Eq.(3), fine cell size d yields high H_{ci}. For this reason, to obtain high H_{ci}, the solution temperature must be raised. But on the other side, higher solution temperature results in larger grain size of magnets and smaller domain wall, which leads to the decrease of domain wall pinning strength and H_{ci}. Therefore, high H_{ci} can be obtained at optimal solution temperature.

2.4 High temperature magnetic properties

The temperature coefficient of coercivity β of Sm(Co, Fe,



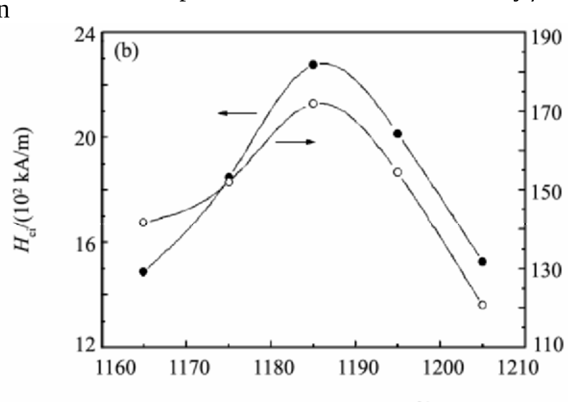


Fig.3 Dependence of magnetic properties on solution temperature of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets
(a) B_r versus solution temperature; (b) H_{ci} and $(BH)_{max}$ versus solution temperature

Cu, $Zr)_z$ magnets between room temperature *RT* and operating temperature *OT* is calculated by Eq.(4):

$$\beta=\frac{H_{ci,RT}-H_{ci,OT}}{H_{ci,RT}\times(RT-OT)}\times100\% \tag{4}$$

This can be rewritten in terms of operating temperature as:

$$OT=RT-\frac{H_{ci,RT}-H_{ci,OT}}{H_{ci,RT}\times\beta} \tag{5}$$

It is safe to set minimum $H_{ci,OT}$=398 kA/m. The maximum operating temperature $(OT)_{max}$ is then determined at $H_{ci,OT}$=398 kA/m[11].

Optimizing the Fe content and processing parameters properly, $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets with high magnetic properties at room temperature can be obtained. To investigate the high temperature magnetic properties of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets, high temperature demagnetization curves are measured, as shown in Fig.4. It is clear that the *B-H* demagnetization curve keeps nearly straight at 500 °C and B_r, $(BH)_{max}$ and H_{ci} at 500 °C reach 0.67 T, 81.2 kJ/m^3 and 509.4 kA/m, respectively, which can be used in such dynamic systems as generator and electric motor and so on. According to Eqs. (4) and (5), β(25–500 °C) 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%/°C and 533 °C, respectively. Compared with commercial Sm(Co, Fe, Cu, $Zr)_z$ magnets, β(25–500 °C) and $(OT)_{max}$ of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnet have been improved effectively. Fig.5 shows the irreversible flux loss for $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets and commercial high coercivity 2:17 type SmCo magnets. The irreversible flux loss of commercial 2:17 type SmCo magnets increase obviously when the temperature is elevated to 400 °C while that of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets increases obviously when the temperature is up to 500 °C, and the irreversible flux loss is 3.78% (length-diameter ratio: *L*/*D*=0.7) at 550 °C, showing good high-temperature stability, which can be used in periodic permanent magnet (PPM) focusing structure[12,13], generator and electric motor and so on.

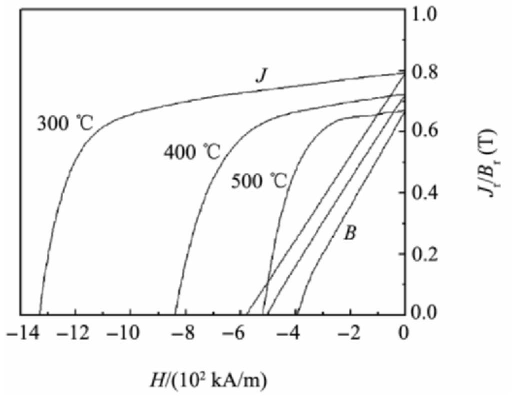


Fig.4 High temperature demagnetization curves of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets

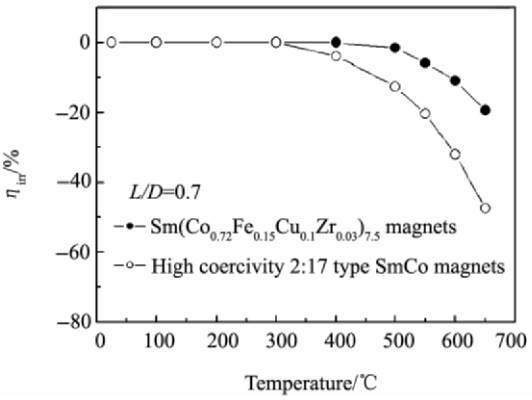


Fig.5 Irreversible flux losse of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets and high coercivity 2:17 type SmCo magnets

3 Conclusion

In summary, high magnetic properties at room temperature could be obtained with proper adjustment of Fe content and optimized processing parameters of $Sm(Co_{bal}Fe_xCu_{0.1}Zr_{0.03})_{7.5}$ (*x*=0.09–0.21) magnets. Higher Fe content brought down the optimal sintering temperature of the magnets, and the optimal Fe content *x* was found to be 0.15. Accordingly the optimal sintering temperature and solution temperature was 1215 and 1185 °C for magnets with *x*=0.15. Prepared by optimal process, B_r, $(BH)_{max}$ and H_{ci} of $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets could reach 0.94 T, 171.9 kJ/m^3 and 2276.6 kA/m at room temperature, respectively. Moreover, $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets exhibited moderately high magnetic properties at 500 °C and good high temperature stability. B_r, $(BH)_{max}$ and H_{ci} at 500 °C reached 0.67 T, 81.2 kJ/m^3 and 509.4 kA/m, respectively, while the temperature coefficient of coercivity β(25–500 °C) reached –0.16%/°C, the maximum operating temperature $(OT)_{max}$ reached 533 °C and the irreversible flux loss was lower than 5% at 550 °C, indicating that $Sm(Co_{0.72}Fe_{0.15}Cu_{0.1}Zr_{0.03})_{7.5}$ magnets would be of great value in high temperature applications.

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