Beneficial effect of Gd substitution on magnetic properties of magnetically anisotropic $SmCo_5$ ribbons

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Magnetically anisotropic $Sm_{1-x}Gd_xCo_5$ ($x=0$, 0.2, 0.5) ribbons are produced by conventional melt spinning. The coercivity H_c increases from 5 kOe for $x=0$ to 11 kOe for $x=0.5$. Microstructure analyses show that this abnormal change of the coercivity is ascribed to the decrease of the quantity of the 2:7 phase, which serves as a center for a reverse domain. The remanence ratio also increases from 0.80 for $x=0$ to 0.95 for $x=0.5$. An energy product of 16 MGOe at room temperature for the $Sm_{0.8}Gd_{0.2}Co_5$ ribbons has been achieved without the need of a complex heat treatment. The temperature characteristic of remanence is substantially improved due to the change of the microstructure arising from Gd substitution.
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Magnetic properties of permanent magnets, and especially their remanence and energy product, are significantly enhanced when they are anisotropic, i.e., the crystallographic *c* axis of different grains of the materials are aligned. Anisotropic permanent magnetic materials with high energy product can be obtained by the process such as powder metallurgy technique,[1–4] directional solidification,[5] and solidification in a magnetic field,[6] but these processes are time and energy consuming. Recently the melt-spinning method is found to be a feasible way of preparing anisotropic permanent magnets.[7–10]

High coercivity in anisotropic $SmCo_5$ ribbons was previously attained only after a complex heat treatment which is believed to optimize the microstructure of the ribbons.[9,10] Our recent experimental results show that high coercivity in anisotropic $SmCo_5$ ribbons was obtained by the introduction of Gd instead of the complex heat treatment. It was found that the coercivity of the $SmCo_5$ ribbons increases with increasing Gd content. Normally the coercivity of $(Sm,Gd)Co_5$ alloys should decrease with the increase of Gd content because Gd substitution leads to a decrease of magnetocrystalline anisotropy field of the $SmCo_5$ phase, which is verified in sintered $Sm_{1-y}Gd_yCo_5$ ($y=0$–0.49) magnets.[11] This abnormal change of the coercivity in the anisotropic $(Sm,Gd)Co_5$ ribbons implies that Gd addition has a different effect on ribbons microstructure with that of sintered magnets. In this letter, the relationship between microstructure and magnetic properties of melt-spun $Sm_{1-x}Gd_xCo_5$ ($x=0$, 0.5) ribbons was discussed in detail.

The alloys with nominal composition $Sm_{1-x}Gd_xCo_5$ ($x=0$, 0.2, 0.5) were prepared by arc-melting. An excess of 15 wt % Sm and 5% Gd was added to compensate for the evaporation. Small pieces of ingot were then inserted into a quartz tube with a nozzle 0.8 mm in diameter. The chamber was evacuated to a vacuum of 10^{-2} Pa and then filled with highly pure argon. The sample was induction melted and ejected through the nozzle using a pressure difference of 0.8 atm. The surface velocity of the Cu wheel was 5 ms^{-1}. Phase components were examined by x-ray diffraction (XRD) with Cu radiation. Microstructural analysis was carried out using a H-9000NA transmission electron microscope (TEM). The ribbons were premagnetized in a field of 20 kOe. Magnetic properties were measured using a Superconduct Quantum Interference Device magnetometer with a maximum magnetic field of 50 kOe at room temperature and a vibrating sample magnetometer with a maximum magnetic field of 8 kOe at high temperature to 300 °C.

Figure 1 shows XRD patterns for as-spun $Sm_{1-x}Gd_xCo_5$ ($x=0$, 0.2, 0.5) alloys. Figures 1(a), 1(f), and 1(d), taken based on the ribbons prepared at $V=5$ m/s clearly show stronger reflections with indices (200) and (110), and the weaker intensity of (111) and (002) in comparison with XRD of the isotropic $SmCo_5$ powder sample [seen in Figs. 1(b), 1(c), and 1(d)]. This indicates that the structure of the ribbons is anisotropic. The *c* axis of crystallites is supposed to be mainly parallel to the longitudinal axis of the ribbon, which is similar to the previous results.[10–12] When compared to the XRD characteristic peaks of power samples of $Sm_{1-x}Gd_xCo_5$ ribbons [see Figs. 1(b), 1(c), and 1(d)], one additional phase, namely of the 2:7 phase which is also observed in the mechanically alloyed $SmCo_5$ and

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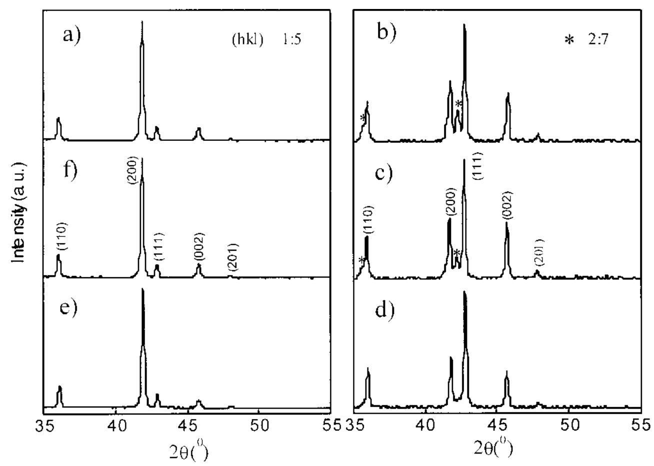

FIG. 1. XRD patterns of $Sm_{1-x}Gd_xCo_5$ [(x=0)(a), 0.2(f), 0.5(e)] ribbons spun at v=5 m/s, the powder of $Sm_{1-x}Gd_xCo_5$ [x=0(b), 0.2(c), 0.5(d)] ribbons spun at v=5 m/s.

$SmCo_5/\alpha$-Fe,[13,14] was detected in the ribbons. It is evident that the relative intensity of diffraction peaks from the 2:7 phase decreases with increasing Gd content. The addition of Gd was found to inhibit the formation of 2:7 phases. Additionally, because of a very stronger diffraction peak intensity of the (200) indice of (Sm,Gd)Co_5 ribbons spun at V=5 m/s, diffraction peaks of 2:7 phase were hardly observed in XRD patterns.

Figures 2(a) and 2(b) show the TEM images revealing the microstructural features of the anisotropic $SmCo_5$ and $Sm_{0.5}Gd_{0.5}Co_5$ ribbons, respectively. These images evidently demonstrate that the microstructure of ribbons depends strongly on the Gd concentration. $SmCo_5$ compound contains a number of the 2:7 phase grains ranging from 100 to 300 nm in sizes as indicated by arrows, which could result in complex defective structure in vicinal areas. Besides, TEM micrographs show that the 2:7 phase also forms a stripe structure as reported previously.[15,16] On the other hand, Gd substitution could apparently improve the quality of the as-spun materials, and note the 2:7 phase has been observed in the $Sm_{0.5}Gd_{0.5}Co_5$ sample in the TEM investigations. These results are in good agreement with the data obtained from XRD measurements. Systematical analyses suggest that Gd substitution restrains the formation of the 2:7 phase.

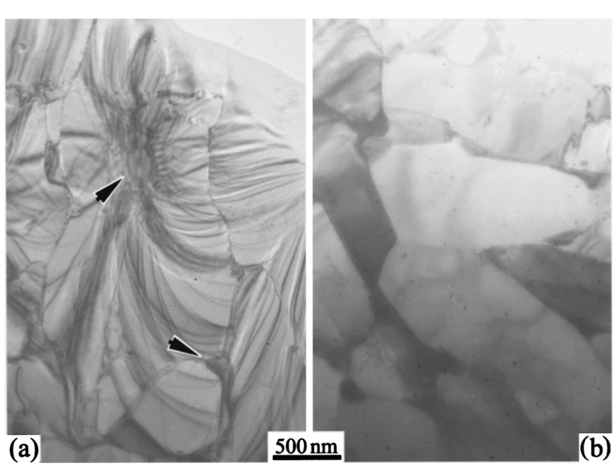

FIG. 2. Comparison of TEM micrographs of anisotropic $Sm_{1-x}Gd_xCo_5$ (x=0, 0.5) ribbons spun at v=5 m/s.

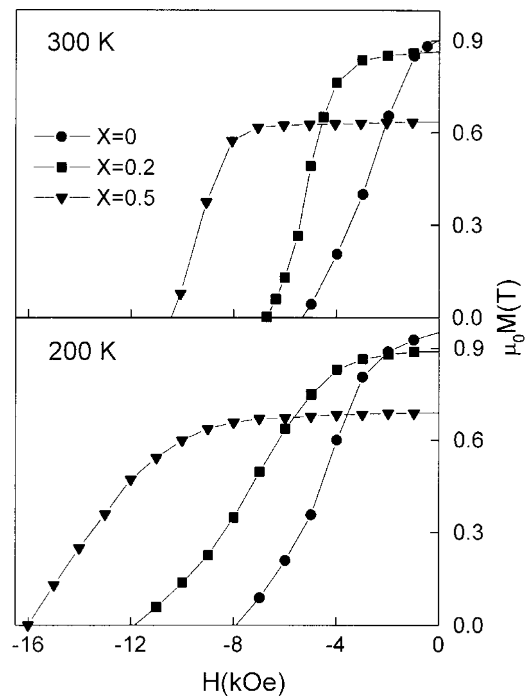

FIG. 3. Demagnetization curves for anisotropic $Sm_{1-x}Gd_xCo_5$ (x=0, 0.2, 0.5) ribbons at 300 and 200 K spun at v=5 m/s.

Demagnetization curves for anisotropic $Sm_{1-x}Gd_xCo_5$ (x=0, 0.2, 0.5) ribbons at 300 and 200 K are shown in Fig. 3. One may note that magnetic properties of the $SmCo_5$ ribbons at 300 and 200 K are greatly affected by the introduction of Gd. Gd substitution leads to a good rectangularity of demagnetization curves and a large coercivity. In the magnetization reversal process, the magnetic moments of the 2:7 phase first reverse in an external field because it has a much lower anisotropy field compared to that of the 1:5 phase, and then the magnetic moments of the 1:5 phase reverse. So the demagnetization curve of $SmCo_5$ and $Sm_{0.8}Gd_{0.2}Co_5$ ribbons contains a shoulder corresponding to a two-step demagnetization behavior which is similar to that observed in decoupled nanocomposite magnets. This behavior at 200 K is more obvious than that at 300 K. Because the 2:7 phase is eliminated by the introduction of Gd, the demagnetization curve of $Sm_{0.5}Gd_{0.5}Co_5$ ribbons at 200 or 300 K presents typical behavior of a single phase. The remanence ratio increases with the increase of Gd content. A high remanence ratio of 0.95 at 300 K has been achieved in the $Sm_{0.5}Gd_{0.5}Co_5$ ribbons. The moderate Gd substitution results in the increase of the energy product at 200 or 300 K. The best energy product at 200 and 300 K are 18 and 16.0 MGOe, respectively.

It was well known that, in order to search high temperature magnets, the important factor is to improve the remanence at high temperature, which is sensitive to the composition of the magnets. The thermal stability of the remanence can be manipulated by optimizing the composition of the magnets. The temperature dependence of remanence for anisotropic $Sm_{1-x}Gd_xCo_5$ (x=0, 0.5) ribbons is shown in Fig. 4. It can be seen that $SmCo_5$ ribbons exhibit temperature coefficient of remanence about -0.09% per °C, and temperature coefficient of remanence for $Sm_{0.5}Gd_{0.5}Co_5$ ribbons is almost $+0.02\%$ per °C among the same temperature range. Generally, the disordering of the rare earth sublattice leads to

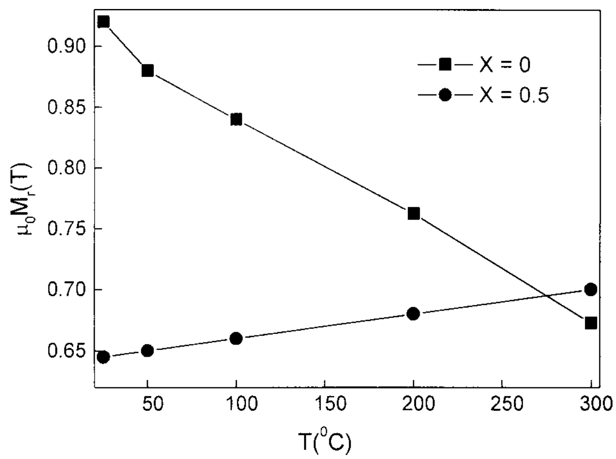

FIG. 4. Temperature dependence of remanence for anisotropic $Sm_{1-x}Gd_xCo_5$ ($x=0$, 0.5) ribbons spun at $v=5$ m/s.

a decrease of magnetization of RCo_5 compounds with increasing temperature, and the ferrimagnetic coupling of heavy rare earth atoms/Co results in an increase of magnetization with increasing temperature in the given temperature range.[17,18] Competition between these two effects decides the temperature dependence of remanence in the given temperature range. In $Sm_{0.5}Gd_{0.5}Co_5$ ribbons, the latter is prior to the former in the range from 25 to 300 °C. So the positive temperature coefficient of remanence for the $Sm_{0.5}Gd_{0.5}Co_5$ ribbons was obtained due to Gd substitution. In addition, the decrease in the amount of the 2:7 phase, which has a lower Curie temperature and saturation magnetization in comparison with that of the $SmCo_5$ phase, is also one of the reasons resulting in the improvement of the temperature coefficient of remanence for $Sm_{0.5}Gd_{0.5}Co_5$ ribbons. In a word, $Sm_{0.5}Gd_{0.5}Co_5$ ribbons will probably become a promising candidate of anisotropic bonded magnets for high temperature applications due to their positive temperature coefficient of remanence.

The beneficial effect of Gd substitution for Sm on magnetic properties of as-spun anisotropic $SmCo_5$ ribbons has been presented. Because Gd addition effectively suppresses the formation of 2:7 phases, anisotropic $Sm_{0.5}Gd_{0.5}Co_5$ ribbons with a high coercivity are produced directly by melt spinning. The remanence ratio increases with the increase of Gd concentration. A high remanence ratio of 0.95 has been attained in $Sm_{0.5}Gd_{0.5}Co_5$ ribbons. The best energy product at 300 and 200 K are 16 and 18 MGOe, respectively. Otherwise, it was found that temperature dependence of remanence of anisotropic $SmCo_5$ ribbons is significantly improved due to Gd substitution. To further optimize magnetic properties of $(Sm,Gd)Co_5$ ribbons, Sm or Gd should be partly replaced by Pr or Nd, etc., which will increase the saturation magnetization of the $SmCo_5$ phase. It is certain that our studies are in favor of simplifying the preparation process, and finally settle a bedrock on volume produce of anisotropic $SmCo_5$ magnets.

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