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Magnetostriction of $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ compounds and their composites ($0.20 \leqslant x \leqslant 0.60$)

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ABSTRACT

The structure, spin configuration, magnetocrystalline anisotropy compensation, magnetic properties, and magnetostriction of $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ ($0.20 \leqslant x \leqslant 0.60$) alloys are investigated. The easy magnetization direction (EMD) at room temperature rotates from the ⟨100⟩ axis ($x \leqslant 0.25$) to the ⟨111⟩ axis ($x \geqslant 0.30$) with increasing Tb content, subjected to the anisotropy compensation between Tb^{3+} and Dy^{3+} ions. The analysis of X-ray diffraction, EMD and magnetostriction show that $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ is an anisotropy compensation system and the compensation point is realized around $x = 0.30$. The Laves phase compound $Tb_{0.4}Dy_{0.5}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ has a large spontaneous magnetostriction, the coefficient λ_{111} up to about 1640 ppm. The epoxy bonded 0–3 and pseudo-1–3 composites were fabricated by curing without and with a magnetic field. A high magnetostriction, longitudinal $\lambda_{||}$ and linear anisotropic $\lambda_a(= \lambda_{||} - \lambda_{\perp})$ up to around 390 and 650 ppm at 6 kOe, respectively, is obtained for the grain ⟨111⟩-oriented pseudo-1–3 epoxy/$Tb_{0.4}Dy_{0.5}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ composite with 20 vol% alloy particles.

1. Introduction

It has been well known that the cubic Laves phase compounds RFe_2 (R = rare earth) exhibit very large anisotropic magnetostriction at room temperature, and the most famous one, $Tb_{0.27}Dy_{0.73}Fe_2$ (Terfenol-D), has been widely used as actuators and transducers [1]. However, the heavy rare earths Tb and Dy are expensive, thus the low-cost materials Pr and Nd have received more attentions for the view point of applications. According to the single-ion model [2], $NdFe_2$ has a large theoretical magnetostriction (about 2000 ppm at 0 K). Besides, the magnetocrystalline anisotropy constant K_1 of both $NdFe_2$ and $DyFe_2$ are positive with the easy magnetic direction (EMD) lying along ⟨100⟩ axis, while the K_1 for $TbFe_2$ is negative with EMD lying along ⟨111⟩ [3]. Thereby, the (Tb,Dy,Nd)Fe_2 should be a good anisotropy compensation alloy system. Recently, the magnetostrictive properties of $Tb_xDy_{0.9-x}Nd_{0.1}Fe_{1.93}$ are reported by Jammalamadaka et al. and found the anisotropy compensation point is near $x = 0.25$, showing it a promising pseudobinary alloy system [4].

Considerable work substituting Fe with Co has been done in an effort to improve the magnetic and magnetostrictive properties of the pseudobinary $(R,R')Fe_2$ alloys [5–10]. It has been pointed out that Co substitution for Fe increases the spin-reorientation temperature of Terfenol-D [5,6] and, Co can improve the formation of Laves phase and suppress the appearance of the non-cubic phase under ambient condition for the alloy system containing light rare earths [7–9]. Zhai et al. [10] found that both Curie temperature and saturation magnetization increase with the slightly increase of Co content for $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ alloys. Recently, we found that Co also has a positive effect on the formation of Laves phase in the $Tb_{1-x}Nd_x(Fe_{0.8}Co_{0.2})_{1.93}$ system [7]. Thus, in the present work, the 20 at.% Co was introduced into $Tb_xDy_{0.9-x}Nd_{0.1}Fe_{1.93}$ system, aiming to develop new magnetostrictive materials for applications.

In case of magnetostrictive composites, these materials are of importance for various applications since they possess the distinct advantages of reducing high-frequency eddy current losses and intrinsic brittleness in comparison with monolithic alloys. In addition, the 1–3 type composite (the embedded phase is connected in one direction and the second phase is connected in all three directions) got more attentions for its excellent anisotropic magnetostriction [11–14]. In this work, the structure, spin configuration, magnetocrystalline anisotropy compensation, magnetic properties, and magnetostriction of $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ compounds are investigated. Spin reorientation was observed with the different Tb/Dy ratio, subjected to the anisotropy compensation between Tb^{3+} and Dy^{3+} ions. The strong ⟨111⟩-oriented pseudo-1–3 type epoxy/$Tb_{0.4}Dy_{0.5}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ composite has been fabricated by curing under a moderate magnetic field. A large linear anisotropic magnetostriction of about 650 ppm for the composite is obtained at an applied magnetic field of 6 kOe.

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2. Experiments

All samples of $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ ($0.20 \leqslant x \leqslant 0.60$) were prepared by arc melting the appropriate constituent metals in a high purity argon atmosphere. The purities of the constituents are 99.9wt% for Tb, Dy and Nd, and 99.8wt% for Fe and Co, respectively. The ingots were sealed in an evacuated quartz tube filled with high-purity argon and homogenized at 670 °C for 7 days, and then furnace cooled to room temperature (RT). X-ray diffraction (XRD) data were recorded at RT with Cu Kα radiation in a D/max-γA diffractometer with a graphite crystal monochromator. To prepare the magnetically aligned samples, the powdered particles of $\leqslant$ 150 μm and epoxy with the weight ratio of 1:2 were homogenously mixed in a plastic mold, and then placed in an electromagnet with a uniform magnetic field of 20 kOe. XRD was implemented on the surface (perpendicular to the curing magnetic field) of the samples, in order to study the easy magnetization direction (EMD) of the Laves phases [14,15]. To investigate the spontaneous magnetostriction λ_{111} of the above compounds, a high-precision XRD step scanning at RT was performed on powdered samples for the (440) peak and then the effect of the $K_{\alpha 2}$ radiation was removed with a standard method [6,16]. Temperature dependence of AC initial susceptibility, X_{AC}, at $H = 2$ Oe, was measured to determine Curie temperatures T_C of the compounds in the alloys. The linear magnetostriction was measured using standard strain-gauge technique in parallel ($\lambda_{||}$) or perpendicular ($\lambda_{\perp}$) direction of applied magnetic fields.

As for the composites, predetermined quantities of alloy particles and epoxy, corresponding to the particles with a volume fraction 20% of the composites, were homogeneously mixed in cubic molds of $10 \times 10 \times 10\ mm^3$. The resulting slurry was degassed under a vacuum for 30 min to eliminate air bubbles. The molds were placed in an electromagnet with and without a uniform magnetic field of 10 kOe along the longitudinal direction of the mold. After aligning and producing chains similar to aligned short-fiber, classified as "pseudo-fiber" configuration (pseudo-1–3 type composite) [17,18], composites were demolded. XRD and magnetostriction were also performed at RT.

3. Results and discussion

The XRD patterns of homogenized $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys, as examples of $x = 0.20$, 0.30 and 0.60, are shown in Fig. 1. It can be seen that all the alloys are essentially the single (Tb,Dy,Nd)$(Fe,Co)_2$ phase with a $MgCu_2$-type cubic structure over the whole concentration range investigated. All lines in the diffraction patterns can be indexed to the characteristics of the Laves phase. The whole X-ray reflection slightly shifts to lower Bragg angles with increasing of Tb content, because of the increase of the lattice constant due to the larger radius of Tb^{3+} ion. The indices (hkl) of the Laves phase are also indexed in Fig. 1.

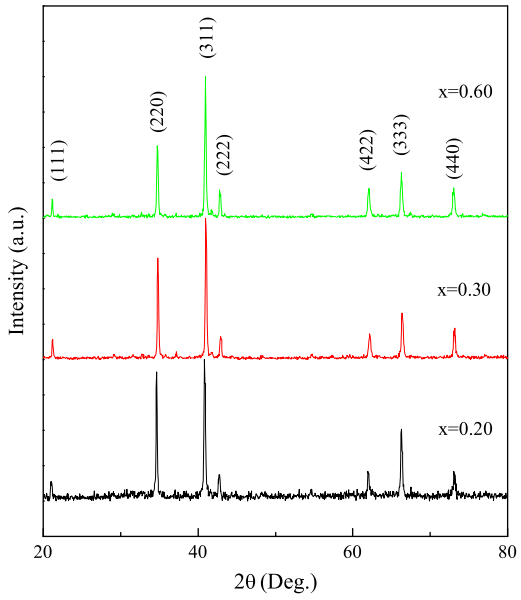

Fig. 1. XRD patterns of the $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys ((*hkl*) of the Laves phase is indexed).

Temperature dependencies of the AC initial susceptibility X_{AC} were measured (the X_{AC}–T curves are not shown here) to determine Curie temperatures T_C of the laves phase in the $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys, and the composition dependence of T_C is shown in Fig. 2. It can be seen that the Curie temperature increases from 685 K to 701 K as x is increased from 0.20 to 0.60, ascribed to that the magnitude of exchange coupling interactions between Tb and Fe/Co atoms is larger than that of Dy–Fe/Co (for $TbFe_2$, $TbCo_2$, $DyFe_2$ and $DyCo_2$, T_C equals 704, 238, 635 and 146 K, respectively) [2,19]. Compared with the Co-free alloy system, the 20 at.% Co substitution for Fe increases T_C and extends their operating temperature scope with about 30 K.

X-ray diffraction patterns at RT of magnetically oriented powders of $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys with $0.25 \leqslant x \leqslant 0.45$ are shown in Fig. 3. As for the samples of $x \geqslant 0.30$, the intensity of (222) peak is strongest accompanied by the strengthened (111) and (333) peaks, which indicates the easy magnetization direction (EMD) lying along ⟨111⟩ direction in accordance with the EMD of Tb is along ⟨111⟩ [4,13]. The (222) peak weakens with decreasing Tb content of $x < 0.30$ and, the (311) peak turns back the strongest one, the same situation as its polycrystalline state, suggesting that the EMD deviates from ⟨111⟩. As for the sample of $x = 0.25$, the (800) peak appears indicating the ⟨100⟩ EMD although the intensity for (800) peak is not the strongest. This incomplete ⟨100⟩ alignment in XRD patterns can be ascribed to that the alloy particles with about 150 μm are not guaranteed to be single domain particles and the curing magnetic field of 20 kOe in preparation process is insufficient to provide the torque needed to rotate the crystallites into alignment completely [15]. As compared with the Co-free $Tb_{0.25}Dy_{0.65}Nd_{0.1}Fe_{1.93}$ compounds where (800) peak can not be observed [4], the 20 at.% Co substitution for Fe increases the spin-reorientation temperature T_{SR} to above RT and slightly changes the composition for the anisotropy compensation to the Tb-rich side at RT. Clark reported that the sign of anisotropy constant K_1 of $TbFe_2$ is negative while K_1 is positive for $DyFe_2$, and the EMD of $TbFe_2$ and $DyFe_2$ lie along ⟨111⟩ and ⟨100⟩, respectively [2]. Thus, the composition dependence of EMD for $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ demonstrates the anisotropy compensation between Tb^{3+} and Dy^{3+} ions and compensation point at RT is around $x = 0.30$ [15,20].

The magnetostriction measurements were performed in a static state with a magnetic field starting from 0 to 10 kOe. The magnetic field dependence of the magnetostriction λ_a ($= \lambda_{||} - \lambda_{\perp}$) for the $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys is shown in Fig. 4(a). It is clear that the saturation is not achieved for all samples. For the samples

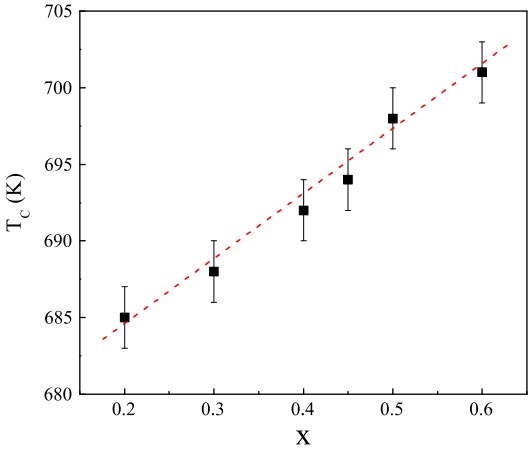

Fig. 2. Composition dependence of Curie temperature T_C of the Laves phase in the $Tb_xDy_{0.9-x}Nd_{0.1}(Fe_{0.8}Co_{0.2})_{1.93}$ alloys.