



Magnetostriction of Laves $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ alloys



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ABSTRACT

Alloys of $\text{Tb}_{0.1}\text{Ho}_{0.9-x}\text{Pr}_x(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ ($0 \leq x \leq 0.50$) are arc melted and investigated for structural, magnetic and magnetoelastic properties. The alloys possess predominantly the cubic Laves phase with MgCu_2 -type structure for $x \leq 0.20$, while impurities are observed for $x \geq 0.30$. The easy magnetization direction (EMD) rotates from $\langle 100 \rangle$ to $\langle 111 \rangle$ axis when x increases from 0 to 0.50, demonstrating the magnetocrystalline-anisotropy compensation between Pr^{3+} and Ho^{3+} ions. The linear anisotropic magnetostriction $\lambda_a = (\lambda_{\parallel} - \lambda_{\perp})$ increases with increasing Pr content when $x \leq 0.30$, ascribed to both the larger magnetostriction of PrFe_2 than that of HoFe_2 and the decrease of the resulted anisotropy due to compensation. The $\text{Tb}_{0.1}\text{Ho}_{0.6}\text{Pr}_{0.3}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ alloy, close to the anisotropy compensation point, has the largest magnetostriction and a low anisotropy at room temperature.

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1. Introduction

There is a great interest in the cubic Laves phase RFe_2 ($R =$ rare earth) compounds in terms of both a fundamental scientific and applied point of view [1–7]. TbFe_2 possesses the largest known magnetostriction at room temperature up to date, and it also exhibits a high anisotropy which is a hindrance for certain applications in acoustic transducers, sensors and actuators, etc. Thus, the anisotropy of TbFe_2 should be minimized by introduction of other R elements while still maintaining a large magnetostriction. The pseudobinary $\text{RR}'\text{Fe}_2$ compounds, e.g. the well-known Terfenol-D ($\text{Tb}_x\text{Dy}_{1-x}\text{Fe}_2$) and $\text{Tb}_x\text{Ho}_{1-x}\text{Fe}_2$, are established as important magnetoelastic materials for their large magnetostriction and low magnetocrystalline anisotropy. According to the prediction of the single-ion model, the theoretical magnetostriction at 0 K of PrFe_2 ($\lambda_{111} \sim 5600$ ppm) is much larger than that of TbFe_2 ($\lambda_{111} \sim 4400$ ppm), DyFe_2 ($\lambda_{111} \sim 4200$ ppm) and HoFe_2 ($\lambda_{111} \sim 1600$ ppm) owing to its large second-order Stevens' factor α_j and average radius squared $\langle r_{4f}^2 \rangle$ of 4f electron shell for Pr^{3+} ion [1]. In addition, the easy magnetization direction (EMD) for PrFe_2 has been reported to be along $\langle 111 \rangle$ axis at room

temperature, while the EMD for TbFe_2 and HoFe_2 is along $\langle 111 \rangle$ and $\langle 100 \rangle$ axis, respectively. It is known that the anisotropy constant K_1 of both TbFe_2 and PrFe_2 is negative, while positive for HoFe_2 . Thus, it is reasonable that the large magnetostriction can be persevered in the $(\text{Tb},\text{Ho})\text{Fe}_2$ system with the introduction of Pr, and the magnetocrystalline anisotropy can be lowered to a minimum by tailoring the composition ratio between Ho and Pr elements. Unfortunately, previous research indicated that it is very difficult to synthesize a single-phase Laves phase $(\text{Tb}, \text{Ho}, \text{Pr})\text{Fe}_2$ alloy containing a high Pr content at ambient pressure, due to the large radius for Pr^{3+} ion [8]. Although many efforts were made to synthesize the Pr-containing alloys by using high pressure and melt-spinning, the complexity of preparing process, the thermal instability, and the residual internal strain in the resulting compounds seriously restrict their potential applications [9,10].

To date, new progress has been made on the synthesis of single Laves-phase of a high Pr content with the addition of stabilizer B/Co [7,8,11,12]. In addition, partial Co substitution for Fe can improve the intrinsic magnetic properties, as an example, increasing Curie temperature and changing spin reorientation temperature (T_{SR}), which significantly impact the EMD and magnetic anisotropy at room temperature (RT). In our recent work, 20 at.% Co has been introduced into $\text{Tb}_{1-x}\text{Ho}_x\text{Fe}_2$ system, and the good magnetostrictive properties were found around the anisotropy compensation point of $x = 0.75$ [13]. In this work, we

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aim to extend our previous work to introduce Pr into the (Tb,Ho)(Fe,Co)₂ system to test the effects of Pr on structure, spin configuration, magnetocrystalline anisotropy and magnetostriction. Here, we choose the ratio of R:(Fe,Co) as 1:1.93 instead of 1:2 in order to obtain a more pure Laves phase [14].

2. Experimental

Polycrystalline Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} alloys ($0 \leq x \leq 0.50$) were prepared by arc melting the appropriate constituent metals in a high-purity argon atmosphere. The purities of the constituents are 99.9 wt.%. The melting was carried out three times to ensure homogenous mixing of the constituents. The ingots were then sealed in quartz tubes in a high-purity argon atmosphere and homogenized at 750 °C for 7 days, and then furnace cooled to RT. X-ray diffraction (XRD) was performed at RT with Cu-K α radiation in a D/max- γ A diffractometer with a graphite crystal monochromator. To prepare magnetically aligned samples, powder particles smaller than 150 μ m were homogeneously mixed with epoxy with weight ratio 1:2 in a plastic mold, and then placed for solidification in a uniform magnetic field of 10 kOe. XRD was carried out on the surface (perpendicular to the direction of the curing magnetic field) of the samples, in order to study the EMD of the Laves phase [7,11]. The magnetization was measured at RT in a vibrating-sample magnetometer (VSM). The two magnetostrictions, including the one due to the application of a parallel applied field H (denoted as $\lambda_{||}$) and the one due to the use of a perpendicular H (denoted as λ_{\perp}), were measured at RT using a metallic strain gauge with a gauge factor of 2.00 and a resistance of 120 Ω attached to the center of the samples and connected to a strain indicator.

3. Results and discussion

The result of powder XRD analysis of the Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} ($0 \leq x \leq 0.50$) alloys indicates the effects of different Pr concentration on the phase composition, as shown in Fig. 1. When $x=0$, one corresponds to the spectrum of (Tb,Ho)(Fe,Co)₂, which is a cubic Laves phase with MgCu₂-type structure and is taken as a reference to compare and verify new phases with

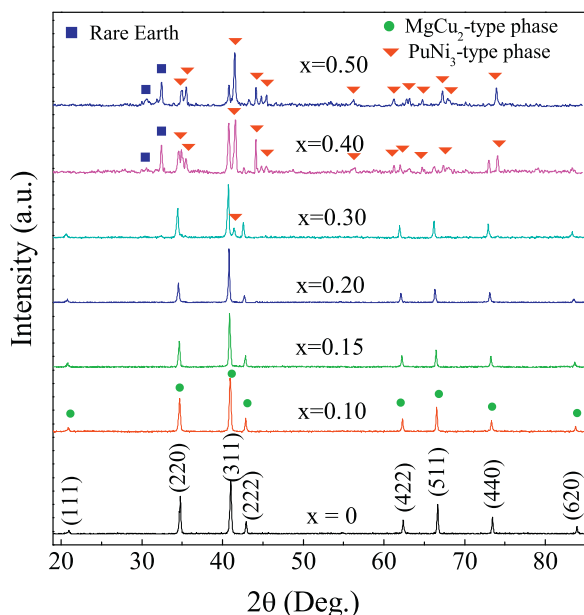


Fig. 1. XRD patterns of the Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} alloys ((h k l) of the Laves phase is indexed).

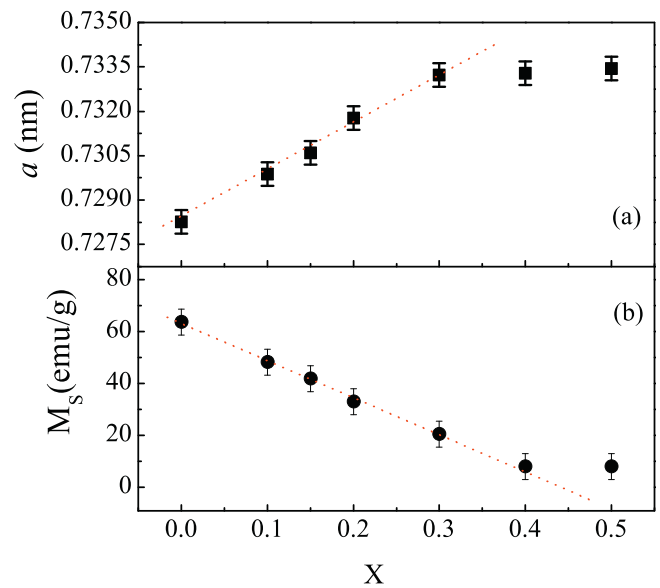


Fig. 2. Composition dependence of (a) the lattice parameter a and (b) the saturation magnetization M_s of the Laves phases in the Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} alloys.

other samples (within the accuracy of XRD). The indices (h k l) of the Laves phase are also indexed in Fig. 1. When Pr content increases with $0.15 \leq x \leq 0.20$, the number of peaks is not change, which indicates that no new phases appear. However, when Pr content increases to be $x=0.30$, new peaks occur, which implies that there is new phase forming. The trace of impurity phase around 41° is detected, which corresponds the (Tb, Ho, Pr)(Fe,Co)₃ phase with PuNi₃-type structure. When Pr content further increases to be $x=0.40$, the rare earth phase appears, coexisting the 1:3 type impurity and the (Tb, Ho)(Fe,Co)₂ Laves phase. As for $x=0.50$, the intensity of (116) peak of the 1:3 type phase becomes the strongest, which indicates that it becomes the main phase. Compared with the Co-free Tb_{0.1}Ho_{0.9-x}Pr_xFe_{1.93} compounds [8], the Pr solubility limit is increase to be 20 at.% in the (Tb,Ho,Pr)(Fe,Co)₂ system, indicating that the partial Co substitution for Fe could help the formation of MgCu₂-type structure with a high Pr content, in good agreement with the previous report [12].

The lattice parameter a of the Laves phase in Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} alloys was calculated from the peaks of XRD spectra, and the dependence of a on Pr content x is plotted in Fig. 2(a). The lattice parameter increases from 0.7282 nm to 0.7332 nm with increasing Pr content up to $x=0.30$, due to the larger radius of Pr³⁺ than of Ho³⁺ ion. It can be seen that the curve $a-x$ approximately follows the linear Vegard's law for $x \leq 0.30$, while deviating the linear trend when $x \geq 0.40$. The linear regularity suggests that the amount of the impurities is small and the secondary phase may be neglected for $x \leq 0.30$. The deviation from linear is due to the emergence of secondary phase such as PuNi₃-type and rare-earth phases, as indicated in XRD patterns (Fig. 1).

The XRD patterns at RT for the magnetically oriented powders of Tb_{0.1}Ho_{0.9-x}Pr_x(Fe_{0.8}Co_{0.2})_{1.93} compounds are shown in Fig. 3. For comparison, the XRD pattern of powders at random state, as an example for $x=0.20$, is also presented in the top panel of Fig. 3. The intensity ratio $I_{(100)}/I_{(311)}$ and $I_{(222)}/I_{(311)}$ vary with changing x , which indicates the magnetocrystalline alignment caused by the variation of magnetocrystalline anisotropy [7]. For the samples with $x \leq 0.15$, the intensity of the (8 0 0) peak is the strongest, while the other weaker peaks appear accordingly. The latter can be ascribed to incomplete alignment, because it is not guaranteed that every particle with about 150 μ m is a single domain particle. Thus,

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