



Optimization on magnetic anisotropy and magnetostriction in $\text{Tb}_x\text{Ho}_{0.8-x}\text{Pr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ compounds



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ABSTRACT

The structure, magnetocrystalline anisotropy compensation, magnetic properties, and magnetostriction of $\text{Tb}_x\text{Ho}_{0.8-x}\text{Pr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ ($0 \leq x \leq 0.30$) polycrystalline alloys have been investigated. X-ray diffraction (XRD) analysis shows that all the alloys stabilize in the single Laves phase with a MgCu_2 -type cubic structure. The lattice parameter, Curie temperature and saturation magnetization monotonically increase with increasing Tb content. The easy magnetization direction (EMD) at room temperature is detected rotating from the $\langle 100 \rangle$ axis ($x \leq 0.10$) to the $\langle 111 \rangle$ axis ($x \geq 0.15$), accompanied by a rhombohedral distortion with large spontaneous magnetostriction coefficients λ_{111} . The analysis of XRD, EMD and magnetostriction shows that $\text{Tb}_x\text{Ho}_{0.8-x}\text{Pr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ is an anisotropy compensation system, and the critical compensation point is realized around $x=0.15$, which shifts to the Tb-poor side compared with the Pr-free counterpart. An optimized effect on magnetostriction especially at a relatively low field ($\lambda_s \sim 445$ ppm, $\lambda_a \sim 510$ ppm/3 kOe) was obtained in $\text{Tb}_{0.15}\text{Ho}_{0.65}\text{Pr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ compound, which is much larger than that of the Pr-free counterpart $\text{Tb}_{0.15}\text{Ho}_{0.85}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ ($\lambda_s \sim 300$ ppm) and the $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$ ($\lambda_s \sim 325$ ppm), due to the 20 at% Pr introduction. Low content of heavy rare earth Tb, low anisotropy, high saturation magnetostriction and large low-field magnetostriction are obtained in $\text{Tb}_{0.15}\text{Ho}_{0.65}\text{Pr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{1.93}$ compound, which may make it a promising magnetostrictive material.

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1. Introduction

RFe_2 (R =rare earth) compounds with C15 Laves structure are interesting from the technological point of view due to their giant anisotropic magnetostriction at room temperature, especially for TbFe_2 , the largest known magnetostriction up to date [1,2]. However, RFe_2 also exhibits high magnetocrystalline anisotropy, which raises large coercive fields and consequently presents a hindrance for practical applications. To minimize the anisotropy and retain a large magnetostriction, pseudobinary $(\text{R}'\text{R})\text{Fe}_2$ is proposed by alloying two different RFe_2 with the same magnetostriction sign but with the opposite signs of the anisotropy constant K_1 . In this way, the well known Terfenol-D ($\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$) and $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$ were constructed, which are now widely used as actuators and transducers. However, the raw materials for heavy rare earths (e.g. Tb, Ho and Dy) are rare and not cost-effective enough. There is a need for developing new advanced magnetostriction materials by introducing low-cost light rare earth, such as Pr and Nd, which

have received much attention for the view point of application [3–5]. Based on the single-ion model, PrFe_2 has a larger theoretical magnetostriction ($\lambda_{111} \sim 5600$ ppm at 0 K) compared with TbFe_2 ($\lambda_{111} \sim 4400$ ppm at 0 K) [1]. Furthermore, the positive first-order magnetocrystalline anisotropy constant (K_1) was deemed for $\text{PrFe}_2/\text{HoFe}_2$, the opposite sign to that of TbFe_2 . Thus, the Pr-containing $(\text{Tb},\text{Ho})\text{Fe}_2$ should be a promising anisotropy compensating alloy system. Unfortunately, previous research indicated that it is very difficult to synthesize a single-phase Laves phase alloy containing a high Pr content at ambient pressure because of the large radius for Pr^{3+} ion. In fact, it remains challenging to prepare Pr-containing compounds with a single Laves phase if the Pr content exceeds 20–25 at% in the rare earth sublattice [6].

Recently, new progress was attained by introducing Co as substitutional atoms into RFe_2 , stabilizing the formation of light rare earth-based C15 structure and improving the intrinsic magnetic properties [3,7–10]. It was reported that the 20 at% Co substitution for Fe could increase Curie temperature T_C and change spin-reorientation temperature, which affects intensively on magnetic anisotropy at room temperature. In our recent work, the anisotropy compensation system $\text{Tb}_{1-x}\text{Ho}_x(\text{Fe}_{0.8}\text{Co}_{0.2})_2$ was

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prepared, and the good magnetostrictive properties were found around the anisotropy compensation point [8]. In this work, we aim to extend our previous work to introduce 20 at% Pr in the (Tb,Ho)(Fe,Co)₂ system to test the effects of Pr on easy magnetization direction (EMD), magnetocrystalline anisotropy and magnetostriction. Here, we choose 20 at% Co substitution for Fe to stabilize Pr elements and set the ratio of R:(Fe,Co) as 1:1.93 instead of 1:2 in order to obtain a more pure Laves phase [10].

2. Experiments

Polycrystalline Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} alloys with $0 \leq x \leq 0.30$ were prepared by arc melting 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 and homogenized at 750 °C for 7 days in a high-purity argon atmosphere and then furnace cooled to room temperature (RT). X-ray diffraction (XRD) data was recorded at RT with Cu-K_α radiation in a D/max-γA diffractometer with a graphite crystal monochromator. To prepare magnetically aligned samples, the powdered particles of $\leq 150 \mu\text{m}$ were homogeneously mixed epoxy with weight ratio of 1:2 in a plastic mold, and then placed for solidification in an electromagnet with a uniform magnetic field of 12 kOe. XRD was implemented 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 [11]. To investigate the spontaneous magnetostriction coefficient λ_{111} , a high-precision XRD step scan of the (440) reflection was performed on powdered samples. Then, the effect of the K_{α2} radiation was removed with a standard method. The magnetization was measured at RT in a vibrating sample magnetometer (VSM). Temperature dependence of ac initial susceptibility χ_{ac} was measured at $H=2 \text{ Oe}$ to determine Curie temperatures T_C of the compounds. Magnetostrictions $\lambda_{\parallel}$ and $\lambda_{\perp}$ of the samples parallel and perpendicular to the applied field were measured with zero applied pre-stress at RT by 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 powder XRD patterns of the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} ($0 \leq x \leq 0.3$) alloys are shown in Fig. 1, which indicates that the homogenized samples are of essentially the single (Tb,Ho,Pr)(Fe,Co)₂ Laves phase, that is, all the diffraction lines can be the characteristic of the cubic MgCu₂-type structure. The indices (h k l) of the Laves phase are also indexed in Fig. 1. It is seen that all the diffraction angles gently shift to lower angles with the increase of Tb content, owing to the larger radius for Tb³⁺ ion compared with Ho³⁺. The lattice parameter a is calculated from the peaks of the XRD spectra and the composition dependence of a is shown in Fig. 2(a). It is obvious that the lattice parameter increases with increasing Tb contents and the curve $a-x$ approximately follows the linear Vegard's law. The linear regularity also suggests that the introduction of the light rare earth Pr element for 20 at% does not affect the single phase of (Tb,Ho)Fe₂ system.

The temperature dependence of ac initial susceptibility χ_{ac} of the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} alloys is shown in Fig. 3. It is seen that the curve $\chi_{ac}-T$ shows only one peak, which corresponds to the Curie temperature T_C of the Laves phase. This is in consistence with the XRD results that the single Laves phase forms, free of any secondary phases. Fig. 2(b) shows the composition dependence of Curie temperature T_C for the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} alloy. T_C

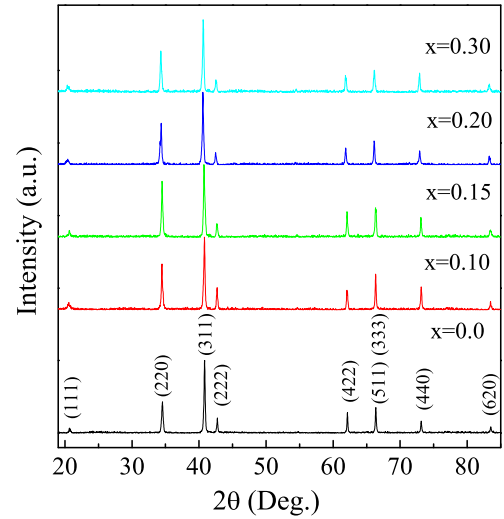


Fig. 1. XRD patterns of the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} alloys ((h, k, l) of the Laves phase is indexed).

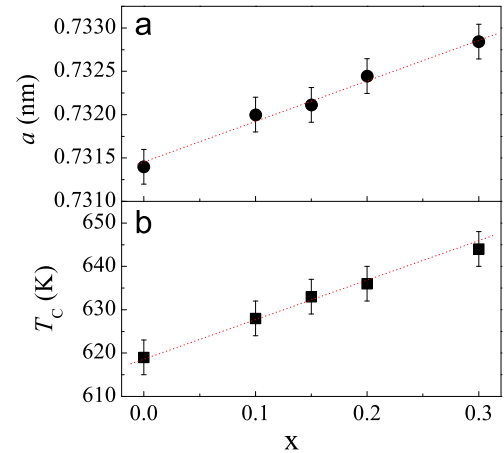


Fig. 2. Composition dependence of (a) the lattice parameter a and (b) the Curie temperature T_C for the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} compounds.

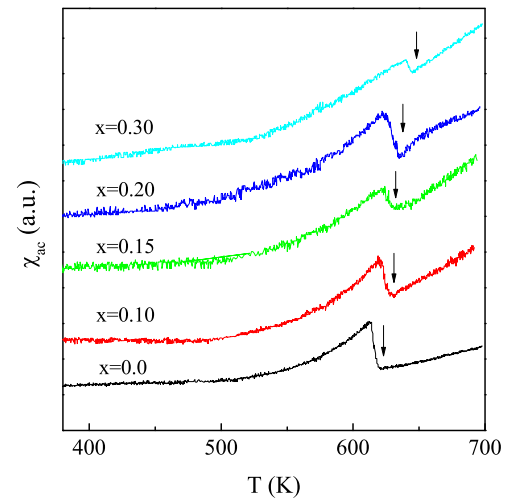


Fig. 3. Temperature dependence of ac initial susceptibility χ_{ac} of the Tb_xHo_{0.8-x}Pr_{0.2}(Fe_{0.8}Co_{0.2})_{1.93} alloys (arrow indicates Curie temperature T_C).

increases from 619 K to 644 K with increasing x from 0 to 0.30, attributed to that the exchange coupling interaction between Tb and Fe/Co atoms is larger than that of Ho-Fe/Co (for TbFe₂, TbCo₂,

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