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Structure and magnetostrictive properties of melt-spun $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ alloys

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ABSTRACT

$Pr(Fe_{0.4}Co_{0.6})_{1.93}$ ribbons were prepared by a melt-spinning method. Their structure and magnetic properties are investigated as functions of wheel speed and annealing temperature. The as-spun ribbon consists of a $Pr(Fe, Co)_2$ cubic Laves phase and an amorphous phase at a wheel speed of $v \geq 35$ m/s, while the non-cubic phases of $PuNi_3$-type and rare earth appear when the speed lower than 30 m/s. A single $Pr(Fe, Co)_2$ phase with $MgCu_2$-type structure has been synthesized by the process for the wheel speed of $v \geq 35$ m/s and subsequent annealing at 500 °C for 30 min. The epoxy/$Pr(Fe_{0.4}Co_{0.6})_{1.93}$ composite has been produced by a cold isostatic pressing technique, and the magnetic properties have been investigated. The composite rod sample possesses good magnetostrictive properties, i.e., a large magnetostriction ($\lambda_a = \lambda_{\parallel} - \lambda_{\perp}$) of 710 ppm at 800 kA/m and a dynamic coefficient d_{33} of 0.67 nm/A at 100 kA/m, and is of practical value.

1. Introduction

The C15 Laves phase RFe_2 (R = rare earth) compounds exhibiting giant magnetostriction attract considerable attention in the last decades [1]. $TbFe_2$ possesses a large magnetostriction ($\lambda_{111} = 2460$ ppm), the largest known one at room temperature (RT) up to date, which is useful in transducers and actuators. However, its high magnetocrystalline anisotropy ($K_1 = -76 \times 10^6$ erg/cm^3) at RT is a hindrance to applications because then large magnetic fields are required to saturate the magnetization and magnetostriction. Unlike $TbFe_2$, $PrFe_2$ is expected to have a larger magnetostriction than that of $TbFe_2$, and the spontaneous magnetostriction coefficient λ_{111} is calculated to be 5600 ppm at 0 K according to the single ion model [1]. Moreover, $PrFe_2$ has a very small anisotropy $K_1 = 7.3 \times 10^6$ erg/cm^3 at 1.5 K, which is even an order smaller than that of $TbFe_2$ at RT [2]. Together with that Pr is much cheaper than Tb, making $PrFe_2$ a promising magnetostrictive candidate for applications at RT. Unfortunately, it is very difficult to synthesize a pure $PrFe_2$ compound with a Laves phase structure under ambient pressure [3]. Many efforts have been paid to the synthesis and the magnetostrictive properties of the pure $PrFe_2$ and the relevant Pr-containing Laves-phase compounds [4–7]. Guo et al. [8] investigated the properties of $Tb_{1-x}Pr_x(Fe_{0.4}Co_{0.6})_{1.9}$ alloys, and found that a remarkable magnetostriction, $\lambda_{\parallel} - \lambda_{\perp} > 800$ ppm at 800 kA/m, is obtained for the $Pr(Fe_{0.4}Co_{0.6})_{1.9}$ alloy even though it contains some amount of $Pr(Fe, Co)_3$ impurities with a $PuNi_3$-type structure. It is possible that the magnetostriction may be improved if $Pr(Fe, Co)_2$ single Laves phase can be obtained while the impurities are eliminated. The non-equilibrium method, melt-spinning with subsequent annealing, was shown to be an effective method to synthesis meta-stable phases which cannot be synthesized at equilibrium condition [9]. Magnetostrictive composites, by integrating magnetostrictive particles with an electrical insulating binder, give more opportunity to broaden the application range, due to the reduction in eddy current loss and the intrinsic brittle nature [10,11]. In this article, $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ alloys were prepared by a melt-spinning method, and the effects of heat treatment on the structure, phase transformation, and magnetic properties were investigated. The quasi-static and dynamic magnetostrictive properties of epoxy/$Pr(Fe_{0.4}Co_{0.6})_{1.93}$ composites, prepared by using a cold isostatic pressing technique, are reported as well. Single-phase $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ compound with cubic $MgCu_2$-type structure forms with proper wheel speed and

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annealing temperature. A good magnetostrictive property, $\lambda_{\parallel}-\lambda_{\perp}=710$ ppm at 800 kA/m and $d_{33}=0.67$ nm/A at 100 kA/m, is obtained for the epoxy/$Pr(Fe_{0.4}Co_{0.6})_{1.93}$ composite.

2. Experiments

Starting ingots having $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ stoichiometry were prepared by arc melting the appropriate constituent metals in a magnetocontrolled arc furnace, which mix the ingredients well with magnetic force, under a high-purity argon atmosphere. The purities of the constituents are 99.9 wt% for Pr, and 99.8 wt% for Fe and Co. The ingot was cut into small pieces of about 3 g, and then homogenized at temperature from 500 to 800 °C for 30 min and seven days in an argon atmosphere, respectively. As for melt-spinning samples, an excess (4 wt%) of Pr over the stoichiometric amount was added to compensate for the mass loss during arc melting and melt-spinning. The ingots were then cut into small pieces of about 10 g, from which the ribbons were fabricated by melt-spinning with a wheel speed over a range from 20 to 40 m/s, followed by annealing at temperature from 500 to 800 °C for 30 min under high-purity argon atmosphere.

X-ray diffraction (XRD) data were recorded at room temperature (RT) with Cu K_{α} radiation in a D/max-γA diffractometer with a graphite crystal monochromator. Measurements for temperature dependence of ac initial susceptibility, χ_{ac}, were carried out at H ($=160$ A/m) to determine the Curie temperatures T_C. The ribbons were crushed into powders to the size less than 78 μm. Then, the powders were mixed with epoxy resin at a proper ratio chosen based on the previous work [12], and the volume fraction was determined to be 0.71. The composites were compacted at a pressure of 100 MPa, and then solidified at freedom state at RT. The composites were demolded and cut into pieces of dimensions $6\times6\times20\,mm^3$. The quasi-static magnetic and magnetostrictive properties of the composites were measured at RT and with zero stress bias using an automated measurement system [13]. Measurements of the magnetic hysteresis loops were conducted by energizing an electromagnet to provide a cyclic magnetic field at 0.1 Hz and measuring the corresponding magnetic flux density B by a search coil wrapped around the samples. The two magnetostrictions, including the one due to the application of a parallel H (denoted as $\lambda_{\parallel}$) and the one due to the use of a perpendicular H (denoted as $\lambda_{\perp}$), were measured using a standard strain gauge technique. The dynamic magnetostriction coefficient d_{33} was measured as a function of the magnetic bias field H_{bias} at a frequency of 1 kHz [14].

3. Results and discussion

The X-ray diffraction (XRD) patterns for the $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ alloys as-cast and heat treated at different temperatures are shown in Fig. 1. It is found that the as-cast sample (Fig. 1(a)) shows three subspectra, corresponding to multiphase structure in the alloy. The main phase is the Pr(Fe, Co)$_2$ Laves phase with a $MgCu_2$-type (1:2) cubic structure, and the secondary phase is the Pr(Fe, Co)$_3$ phase with a $PuNi_3$-type (1:3) structure, coexisting with a small amount of rare earth phases. They are also indicated with symbols indexed in Fig. 1. Upon heat treatment at different temperatures, the phase characteristics are almost the same as the as-cast sample, indicating they are of three-phase structure. As an example, the as-cast samples were annealed for 30 min at the temperatures varied from 500 to 800 °C, and the XRD patterns (Figs. 1(b–d)) do not change except for the diffraction intensity, which shows the three phases still existing. To study the annealing time effect on the phases, the as-cast samples were annealed between 500 and 800 °C with the prolonged time of 1–7 d, and the secondary phase Pr(Fe, Co)$_3$ impurity cannot be eliminated (the XRD patterns is not shown here). These results are in good agreement with that reported by Guo [8], indicating that the Pr(Fe, Co)$_2$ single Laves phase cannot be formed by annealing as-cast sample.

The XRD patterns for the $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ ribbons quenched at various wheel speeds are shown in Fig. 2. It can be seen that the main phase is the cubic Laves phase for all the ribbons. As for the samples fabricated at relatively high wheel speeds ($\geq$35 m/s), the XRD patterns show some broad crystalline peaks and a broad hump, indicating a mixture of ultra-fine grains of the main Pr(Fe, Co)$_2$ Laves phase and an amorphous phase. There is a small amount of impurities with Pr(Fe, Co)$_3$ and rare earth phases for the samples fabricated at relatively low wheel speeds ($\leq$30 m/s), and especially obvious at the speed of 20 m/s. But, the peak corresponding to the Pr(Fe, Co)$_3$ impurity cannot be observed and

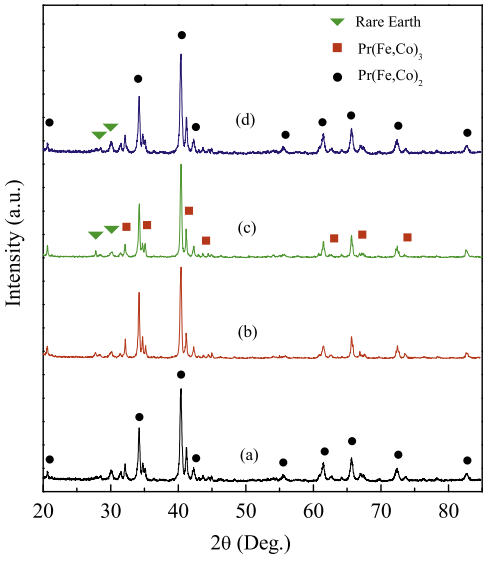

Fig. 1. X-ray diffraction patterns of (a) as-cast and (b–d) post-annealed for 30 min at 500, 700 and 800 °C, respectively, of the $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ alloys.

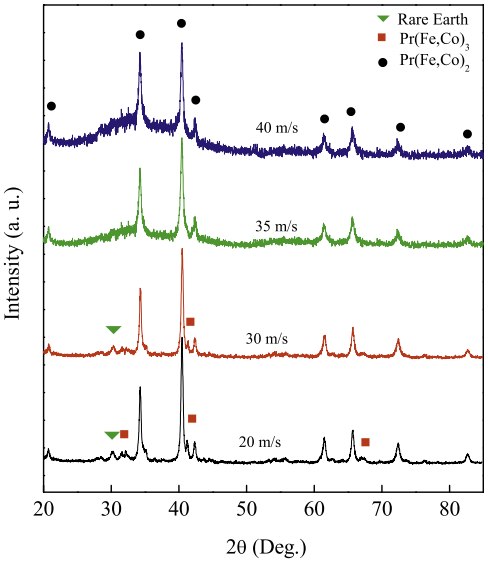

Fig. 2. X-ray diffraction patterns of the as-spun $Pr(Fe_{0.4}Co_{0.6})_{1.93}$ alloys at different speeds for 20, 30, 35 and 40 m/s, respectively.