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Structure and magnetic properties of melt-spun $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ compound

Z.Y. Chen [a], Y.G. Shi [a,*], L. Wang [a], Q. Pan [a], H.F. Li [b], D.N. Shi [a]

[a] Department of Applied Physics, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China
[b] State Key Laboratory of Mechanics and Control of Mechanical Structures, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

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ABSTRACT

High-Nd content $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ cubic Laves compound which could not be prepared by annealing its as-cast ingot was successfully fabricated by melt-spinning and subsequent low-temperature annealing. The effects of wheel speed and annealing temperature on the structure and magnetic properties of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ ribbons are investigated. The ribbons with single cubic Laves phase were obtained at a wheel speed of 40 m/s and subsequent annealing temperature of 773 K. The average grain size decreases with the increase of the wheel speed from 10 m/s to 40 m/s. The easy magnetic direction of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ Laves phase lies along [111] at room temperature, which was confirmed by Mössbaure spectra. Meanwhile, the resin-bonded ribbons obtained at a wheel speed of 40 m/s have a magnetostriction of 321 ppm at room temperature. This work demonstrates an alternate way to synthesize the metastable high-Nd content cubic Laves phase magnetostrictive compounds under ambient pressure other than high-pressure annealing.

1. Introduction

The C15 cubic Laves RFe_2 (R = rare earth) magnetostrictive compounds, such as, Terfenol-D ($Tb_{0.27}Dy_{0.73}Fe_2$) and $Tb_{0.15}Ho_{0.85}Fe_2$, have been widely applied in sonar transducer, sensors and actuators, owing to their function of converting electrical energy to mechanical energy [1,2]. Considering the much richer mineral resources of Nd than heavy rare earths and large theoretic magnetostriction of $NdFe_2$ (2000 ppm at 0 K [1]), much attention has been paid to the synthesize and study the magnetostrictive properties of Nd-contained cubic Laves compounds in the past few years [3–10]. The same with $HoFe_2$ and $DyFe_2$, the easy magnetic direction (EMD) of $NdFe_2$ lies along [100] at room temperature [11]. Therefore, $Tb_{1-x}Nd_xFe_2$ should also be a promising anisotropy compensation system, like $Tb_{1-x}Dy_xFe_2$ and $Tb_{1-x}Ho_xFe_2$. Unfortunately, the radius ratio of Nd:Fe is too large to fit the ideal atomic radius ratio for cubic Laves phase compound ($\sqrt{3/2}$ [12]), precluding the ambient pressure synthesis of $Tb_{1-x}Nd_xFe_{1.9}$ cubic Laves phase when the Nd concentration is higher than 60 at% in the rare earth sublattice [4]. Therefore, one of the key aspects in the research of Nd-containing magnetostrictive materials is how to synthesize single cubic Laves compounds with high-Nd content. Ren et al. [3] found that the introduction of B can improve the content of Nd in $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$. However, the single Laves phase was obtained only when $x \leq 0.55$. Our group [7] recently reported the structure and magnetostrictive properties of $Nd(Fe_{1-x}Co_x)_{1.9}$ compounds. It was found that the substitution of Co for Fe is beneficial for the synthesis of $Nd(Fe_{1-x}Co_x)_{1.9}$ cubic Laves compound. However, single cubic Laves phase can be obtained only when Co content up to 60 at% in the transition metal sublattice by a traditional annealing method under normal pressure. Meanwhile, both the Curie temperature and saturation magnetization of $Nd(Fe_{1-x}Co_x)_{1.9}$ increase with increasing Co concentration to a maximum at $x = 0.2$. Recently, Yin et al. [8] investigated the structure and magnetostrictive behavior of $Tb_{1-x}Nd_x(Fe_{0.8}Co_{0.2})_{1.93}$ compounds prepared by annealing as-cast ingots under ambient pressure. It was found that non-cubic phase appears when $x \geq 0.4$. Shi et al. [4] have successfully synthesized high-Nd content cubic Laves $Tb_{1-x}Nd_xFe_{1.9}$ anisotropy compensation compounds by arc-melting and subsequent high-pressure annealing. As a matter of fact, alternative methods of synthesis high-Nd content RFe_2 magnetostrictive materials without involving the high-pressure process are still highly desired. The rapid quenching by melt-spinning was reported to be an effective way to synthesize

* Corresponding author.
E-mail addresses: shiyangguang@gmail.com, shiyg@nuaa.edu.cn (Y.G. Shi).

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metastable phases [13–15]. As far as known, few investigations on synthesizing and stabilizing high-Nd content cubic Laves compounds by melt-spinning technique were reported.

In the present work, we have prepared high-Nd content $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ compound with single cubic Laves phase by melt-spinning and subsequent annealing. The structure, magnetic properties and magnetostriction of the compound were investigated. This work may provide an effective way to synthesize high-Nd content cubic Laves phase under ambient pressure.

2. Experiment

Starting ingots of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ stoichiometry were melted by an arc furnace with constituent metals under high purity Ar atmosphere. The purity of constituent metal is 99.9% for Tb and Nd and 99.99% for Fe and Co. Extra 5 wt.% of Tb and Nd were added to compensate the loss during the arc-melting and melt-spinning. For a better chemical homogeneity, the ingots were remelted four times. Subsequently, the ingots were induction melted and then spinning at different wheel surface speeds from 10 m/s to 40 m/s under Ar atmosphere. Conventional X-ray diffraction (XRD) analysis was carried out using Cu K α radiation with a Rigaku D/MaxgA diffractometer at room temperature. The ribbons were then vacuum annealed at different temperature from 773 K to 973 K for 30 min and cooled with the furnace. The atomic force microscopy (AFM) was performed with a commercial scanning probe microscope (Digital Instruments, NanoScope V, Veeco). The magnetization of the compounds was measured using a vibrating sample magnetometer (VSM) at room temperature. The temperature dependence of magnetization was measured at a field of 2 kOe by VSM. In order to detect the easy magnetic direction of the sample, the ^{57}Fe Mössbauer spectra were recorded at room temperature, calibrated with a standard α-Fe foil and analyzed by Lorentzian lines in 256 channels using software MossWinn [16]. To measure the magnetostriction of the sample, annealed ribbons were crush into powders and mixed with 5% epoxy resin and pressed to 60 MPa to produce cylindrical disk with size $\phi 10mm \times 2mm$. The linear magnetostriction was measured using standard strain-gauge technique in directions parallel ($\lambda_{\parallel}$) or perpendicular ($\lambda_{\perp}$) to applied magnetic fields at room temperature.

3. Results and discussion

Fig. 1 shows the XRD patterns of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ as-cast ingots and annealed at different temperature for 7 days. It can be seen that each sample exhibits a multiphase structure. For the as-cast ingot [Fig. 1](a), the main phase is (Tb,Nd) $(Fe,Co)_3$ with the $PuNi_3$-type structure, and the second phase is (Tb,Nd) $(Fe,Co)_2$ with $MgCu_2$-type structure, coexisting with a small amount of rare earth phases. The XRD patterns of the ingots annealed at 773 K [Fig. 1](b) and 973 K [Fig. 1](c) are similar with the as-cast one. Here, (Tb,Nd) $(Fe,Co)_3$ remains the dominate phase and (Tb,Nd) $(Fe,Co)_2$ is the second one. From these results, we can find that it is difficult to synthesize $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ with single cubic Laves phase by annealing its as-cast ingots under normal pressure, which should be ascribed to the large radius of Nd^{3+} [12]. Shi et al. investigated the structure of $Tb_{1-x}Nd_xFe_{1.9}$ compounds annealed under ambient pressure. However, no trace of cubic Laves phase with $MgCu_2$-type structure can be found in their samples when x≥ 0.6 [4]. Here, the emergence of cubic Laves phase should be attributed to the effects of substitution of Co for Fe, which is consistence with the previous reports [7,8].

Fig. 2 presents the room-temperature $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ XRD patterns of as-spun ribbons at a wheel speed of 40 m/s and annealed at 773 K and 973 K for 30 min, respectively. In contrast

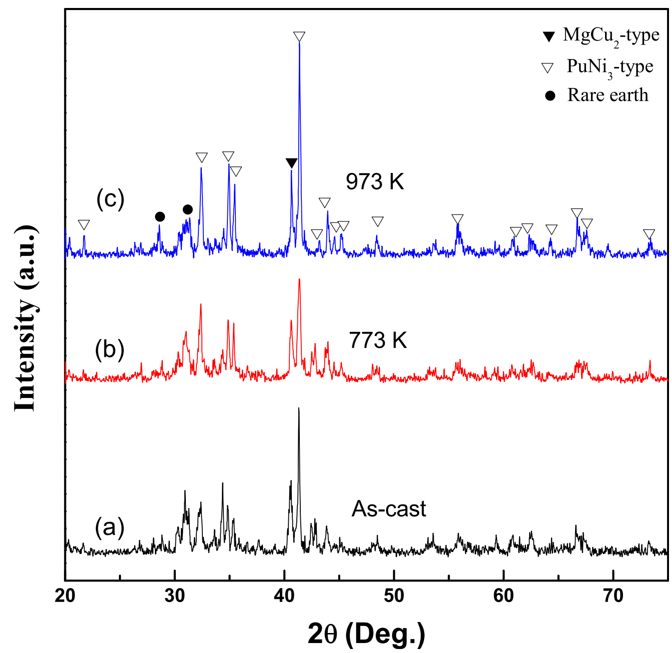

Fig. 1. The room-temperature XRD patterns of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ compound: (a) as-cast ingots and annealed at (b) 773 K and (c) 973 K for 7 days.

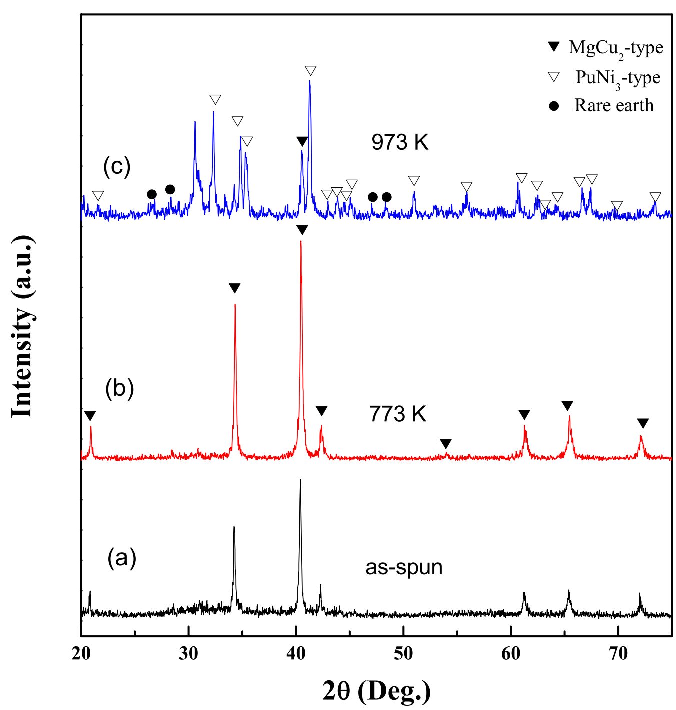

Fig. 2. The room-temperature XRD patterns of $Tb_{0.2}Nd_{0.8}(Fe_{0.8}Co_{0.2})_{1.9}$ ribbons: (a) as-spun at wheel speed of 40 m/s and annealed at (b) 773 K and (c) 973 K for 30 min.

with Fig. 1, both the ribbons of as-spun and annealed at 773 K are free of $PuNi_3$-type structure and exhibit almost single cubic Lave phase with $MgCu_2$-type structure, which indicates that rapid quenching is beneficial for the formation of cubic Laves phase and can eliminate the (Tb,Nd) $(Fe,Co)_3$ phase effectively. This also means that high-Nd content single Laves compounds, which could not be readily synthesized by annealing its ingots, can be