Enhanced Magnetostriction of a Narrow Hysteresis

Tb_{0.26}Dy_{0.54}Ho_{0.20}Fe₂ Alloy

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[Manuscript received 5 February 2013, in revised form 17 April 2013]

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In this work, a magnetic annealing method used to enhance the magnetostrictive property of a narrow hysteresis alloy Tb_{0.26}Dy_{0.54}Ho_{0.20}Fe₂ is reported. Cylindrical-rod shaped specimen with <110> crystal orientation was fabricated using zone-melting unidirectional solidification technique, followed by annealing in a transverse magnetic field of 366 kA/m. The crystal orientation and bi-phase solidified morphology can be retained after magnetic annealing. A high magnetostriction of 1.508×10^{-3} was obtained in the magnetically annealed specimen, which is 25.2% larger than the untreated one. Simultaneously, the magnetostriction hysteresis width is slightly enlarged from 4.45 to 6.36 kA/m, which is still much lower than that of the Ho-free Tb_{0.3}Dy_{0.7}Fe₂ alloy. The additional anisotropy which is induced by magnetic annealing, as reflected by the magnetic hysteresis loops, is responsible for the enhancement of magnetostrictive performance.

KEY WORDS: Magnetostriction; Magnetostriction hysteresis; Induced anisotropy

1. Introduction

Because of the important applications in actuators, sensors and transducers, TbDyFe alloy has attracted considerable interest as a kind of well-known giant magnetostrictive material (GMM)^[1–3]. To meet with the requirements of high magnetostrictive performance under modest external magnetic fields, <112> and <110> oriented polycrystals have been fabricated by various unidirectional solidification techniques^[4,5]. When these oriented polycrystals were subjected to a uniaxial compressive pre-stress, they will show a dramatic magnetostriction improvement, which is the so-called “jump” effect^[6]. However, the presence of pre-stress enlarges the nonlinear hysteresis during magnetization-demagnetization cycles, which has been deemed as one major concern for many practical applications because the accompanied heat or energy loss could be quite large. One efficient approach to solve this problem is adding a third rare earth ele-

ment, such as Ho, minimizing both of the two magnetocrystalline anisotropy constants, K_1 and K_2 simultaneously^[7]. Therefore, a series of Ho-containing alloys, such as Tb_{0.20}Dy_{0.22}Ho_{0.58}Fe_{1.95}, Tb_{0.28}Dy_{0.57}Ho_{0.15}Fe_{1.95}^[8] and Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe₂^[9] have been synthesized, exhibiting much lower hysteresis. For example, a <112> oriented Tb_{0.28}Dy_{0.57}Ho_{0.15}Fe_{1.95} possesses a hysteresis width of only 10.74 kA/m, which is 15% lower than the Ho-free alloy Tb_{0.3}Dy_{0.7}Fe_{1.95}^[8]. Unfortunately, the magnetostrictive performance is drastically deteriorated because Ho has only 1/3 of the intrinsic magnetostriction of Tb and Dy^[1]. Therefore, it is of importance to improve the magnetostrictive performance of the narrow hysteresis TbDyHoFe GMM. Previous investigations demonstrate that magnetic annealing is a useful tool to improve the magnetostriction of TbDyFe oriented polycrystals^[10–12]. It seems that this method has a better effect for the alloy with a larger magnetocrystalline anisotropy constant K_1 . Verhoeven *et al.*^[11] reported that axial magnetostriction of the <112> oriented Tb_{0.32}Dy_{0.68}Fe_{1.96} can be improved dramatically from 8.0×10^{-4} to 1.99×10^{-3} when it was annealed under a transverse external

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field. Our recent work^[12] showed that the magnetic annealing can generate a magnetostriction as high as 2.68×10^{-3} in $\langle 110 \rangle$ oriented polycrystals $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.95}$, which possesses a relatively smaller K_1 than $\text{Tb}_{0.32}\text{Dy}_{0.68}\text{Fe}_{1.96}$. The purpose of this work is whether the magnetic annealing is effective or not to enhance the magnetostrictive property of narrow hysteresis Ho-containing alloys, whose magnetocrystalline anisotropy constants K_1 and K_2 are nearly equivalent to zero. We conducted transverse magnetic annealing on a $\langle 110 \rangle$ oriented alloy with a much lower K_1 than $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.95}$ and investigated the evolutions of crystal orientation, morphologies, magnetostrictive performance and hysteresis width after magnetic annealing.

2. Experimental

Master alloy with a nominal composition of $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ was prepared by arc melting method by using Tb, Dy, Ho and Fe with purity of 99.9% as the raw materials in a vacuum furnace. By using a water-cooled copper mould, an ingot was then cast with a diameter of 6.8 mm. The cylinder rod was directionally solidified by zone-melting method with a growth velocity of 480 mm/h. A $\langle 110 \rangle$ preferred crystal orientation was formed along the rod axis, which was identified by X-ray diffraction (XRD) measurement. From the thermal gravity curve (as shown in Fig. 1) measured under a static magnetic field^[13], Curie temperature was determined to be 368.6 °C. The oriented rod (7 mm in diameter and 25 mm in length) was then heated to 500 °C in a vacuum chamber, which is appropriately 130 °C higher than T_C , and kept for 10 min. An external field of 366 kA/m was then applied perpendicular to its axis during it was cooled to the room temperature^[14]. Longitudinal magnetostriction $\lambda_{//}$ was measured by a standard strain gauge method under a series of uniaxial compressive pre-stresses. To initialize each measurement, the specimen was magnetized up to 560 kA/m

Fig. 1 Thermal gravity curves of $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ measured under a static magnetic field, the arrow reflects the Curie transition

and then slowly demagnetized to 0 kA/m. Both the compressive pre-stress and magnetic field were applied along the rod axis. For the aspect ratio of the specimen is larger than 3:1, the demagnetization field can be neglected here. Magnetic hysteresis loops for both untreated and magnetically annealed samples were conducted by a lakeshore 7407 vibrating sample magnetometer (VSM). To determine the spontaneous magnetostriction constant λ_{111} from lattice distortion, high-resolution synchrotron XRD was employed to observe the splitting diffraction peaks. The patterns for powder samples were measured by the BL 15XU NIMS beam line of Spring-8, which can provide a strain resolution of about 5×10^{-4} ^[15]. The X-ray wavelength is 0.063 nm.

3. Results and Discussion

3.1 Crystal orientation and morphology

Room temperature XRD patterns recorded with $\text{CuK}\alpha$ radiation for polycrystalline powders, transverse sections of the unidirectionally solidified and magnetically annealed $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ specimens were shown in Fig. 2. The polycrystalline samples (Fig. 2(a)) can be identified as MgCu_2 -type Laves phase with cubic structure, where (311) peak is the strongest one. However, the intensity of (311) peak became weaken while those for the (220) and (440) peaks were enhanced for the unidirectionally solidified and magnetically annealed specimens. In addition, (220) peak became the strongest one, which indicates that $\langle 110 \rangle$ is the preferred orientation along the rod axis during unidirectional solidification and retained after magnetic annealing.

Fig. 3 shows typical morphologies of the $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ rods before and after magnetic annealing. Both specimens exhibit similar bi-phase structure, which is similar to the oriented TbDyFe alloys^[16]. The specimen consisted of

Fig. 2 XRD patterns of $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ powders (a), the transverse sections of as-directionally solidified (b) and magnetically annealed (c) $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ rod

Fig. 3 Optical micrographs for the transverse sections of as-directionally solidified (a) and magnetically annealed (b) $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ rods

the plate-like Laves phase (gray matrix) and the net-shaped RE-rich phase (black regions) which is distributed at the boundaries, respectively. No obvious change occurred in the morphology after magnetic annealing because the annealing temperature is much lower than the eutectic reaction temperature in Tb-Dy-Fe system^[17].

3.2 Magnetostriction and hysteresis

Magnetostrictions of the $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ alloy under a series of compressive pre-stresses are shown in Fig. 4. The inset shows the stress magnitude. The magnetostriction jump effect is obvious for both specimens, which is a typical feature of TbDyFe oriented polycrystal. For instance, saturation magnetostriction (λ_s) of the untreated specimen can be enhanced from 7.0×10^{-4} to 1.204×10^{-3} with the application of a pre-stress of 30 MPa. As proposed by Clark *et al.*^[6], the magnetostriction jump effect is ascribed to the stress induced magnetization realignment along the perpendicular direction (perpendicular to the rod axis). Such realignment will favor the 90° magnetization rotation, which can generate much larger magnetostriction. Simi-

larly, the initial realignment of magnetization can also be realized by magnetic field annealing because the reorientation of magnetization is indeed easy due to the paramagnetic-ferromagnetic transition around T_C . As a result, the magnetic annealing introduces extra anisotropy through redistributing the domain magnetizations and consequently changes the magnetization rotation process. In the present work, after magnetic annealing, saturation magnetostriction under free conditions (without applying any stress) can increase from 7.0×10^{-4} to 8.75×10^{-4} . When a compressive pre-stress was subjected, magnetostrictive performance of the magnetically annealed specimen could be further enhanced. Under 30 MPa, the saturation magnetostriction can reach 1.508×10^{-3} , which is comparable to the reported data in $\langle 110 \rangle$ oriented $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.95}$ (1.68×10^{-3})^[18].

By deviating the magnetostriction curves, the magnetostrictive strain coefficient $d_{33} = d\lambda/dH$ (where λ is the magnetostriction, H is the magnetic field intensity) as a function of magnetic field were obtained, as shown in Fig. 5. For each case, d_{33} first reaches a peak and then decreases with further increasing external field. Generally, maximum d_{33} could be obtained when the 71° domain wall energy minimizes at this critical magnetic field^[19,20]. Simultaneously, increasing the pre-stress magnitude shifts the peak position toward higher field regions. Additionally, maximum d_{33} will also increase after magnetic annealing, which also reflects the induced extra anisotropy. For example, maximum d_{33} under 0 MPa is only 10.22 nm/A but increases to 13.46 nm/A after magnetic annealing.

The magnetostriction hysteresis width, W_h , is defined as the gap between the increasing and decreasing magnetostriction curve segments at the center point of the magnetostriction change, $\Delta\lambda$, located at the average field where d_{33} reaches a maximum^[18]. The stress magnitude dependence of W_h is shown in Fig. 6(a). For the untreated specimen, W_h initially increases from 0.32 kA/m under 0 MPa to 5.40 kA/m under 25 MPa, and then decreases to 2.86 kA/m under 50 MPa. The hysteresis is ascribed to the irreversible rotation

Fig. 4 Magnetostriction curves for $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$: (a) as-directionally solidified; (b) magnetically annealed

Table 1 The magnetocrystalline anisotropy constant K_1 , magnetostriction hysteresis W_h , magnetostriction λ_s and the spontaneous magnetostriction λ_{111} of the $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ alloy with different states, compared with the reported $\text{Tb}_{0.29}\text{Dy}_{0.48}\text{Ho}_{0.23}\text{Fe}_2$ and $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ alloys

Alloy	K_1 (10^4 J/m^3)	W_h at 0 MPa (kA/m)	λ_s at 0 MPa (10^{-4})	W_h at 30 MPa (kA/m)	λ_s at 30 MPa (10^{-3})	λ_{111} (10^{-3})
$\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ ^[18]	−6.0	6.76 ^[21]	9.40 ($0.57\lambda_{111}$)	9.57 ^[21]	1.680 ($1.02\lambda_{111}$)	1.640
$\text{Tb}_{0.29}\text{Dy}_{0.48}\text{Ho}_{0.23}\text{Fe}_2$ ^[22]	−5.2	4.7	7.37 ($0.48\lambda_{111}$)	8.04 ^[9]	1.463 ^[9] ($0.95\lambda_{111}$)	1.543
$\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$	−3.6	0.32	7.00 ($0.50\lambda_{111}$)	4.45	1.204 ($0.86\lambda_{111}$)	1.393
$\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ (magnetic annealing)	$K_1 + K_u$	1.91	8.75 ($0.63\lambda_{111}$)	6.36	1.508 ($1.08\lambda_{111}$)	1.393

Fig. 5 Magnetostrictive strain coefficient d_{33} vs. magnetic field intensity for as-directionally solidified (a) and magnetically annealed (b) $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ specimens

of the magnetizations toward the perpendicular plane. Under a certain compressive stress, the magnetization rotation changes from irreversibility into reversibility, thus a decreased hysteresis can be observed^[18]. The magnetically annealed specimen shows similar stress dependence to the untreated one. The only difference between them is that maximum W_h occurs under a higher critical pre-stress for the magnetically annealed one. Maximum W_h increases from 5.40 kA/m under 25 MPa for the untreated specimen to 6.68 kA/m under 35 MPa for the magnetically an-

nealed one. Stress dependences of λ_s for $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ are shown in Fig. 6(b). The increase of magnetostriction with stress magnitude reflects the magnetostriction jump effects for both specimens.

Table 1 summarizes K_1 , W_h and λ_s at 560 kA/m under different pre-stresses of $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$, $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ and $\text{Tb}_{0.29}\text{Dy}_{0.48}\text{Ho}_{0.23}\text{Fe}_2$ ^[9], respectively. It should be noted that W_h for the untreated $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ under 30 MPa is only 4.45 kA/m, which is much lower than that for $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ (9.57 kA/m)^[21]. It is demonstrated that Ho addition does provide a drastic reduction in magnetostriction hysteresis. After magnetic annealing, in addition to the significant enhancement of λ_s , W_h just increases slightly to 6.36 kA/m, which is still lower than $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$. It indicates that the narrow hysteresis is still maintained for our magnetically annealed specimen.

From the perspective of energy, the theoretical model of magnetostriction hysteresis W_h is proposed as^[22]:

$$W_h = \alpha \frac{2|K_1|}{\mu_0 M_s} \quad (0 \leq \alpha \leq 1) \quad (1)$$

where α is orientation factor determined by the direction of external field and the orientation of the initial magnetic domain moment, K_1 is the magnetocrystal-

Fig. 6 Stress dependences of hysteresis width (a) and saturation magnetostriction (b) for $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$

Fig. 7 Magnetic hysteresis loops for as-directionally solidified and magnetically annealed specimens

line anisotropy constant, and $\mu_0 M_s$ is the saturation magnetization. Fig. 7 illustrates the magnetic hysteresis loops for $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ both before and after magnetic annealing, which can explain well the change of magnetostriction hysteresis after magnetic annealing by the formula above. The transverse field annealing results in not only an increment in the area of hysteresis loops, but also a shift of the curve toward higher field regions, which is another evidence of the larger anisotropy because of the induced additional anisotropy by magnetic annealing^[10]. In the previous work^[23], it was also suggested that the induced additional anisotropy tends to switch the magnetizations align along one or two principal easy axes, forming a uniaxial anisotropy, which is beneficial to the improvement of magnetostrictive performance. Therefore, the enlargement of magnetocrystalline anisotropy constant will play a dominant role in the increment of hysteresis field, as shown in Fig. 6(a).

According to the single- $\{111\}$ -twin growth model proposed by Jiang *et al.*^[24], saturation magnetostriction of the $\langle 110 \rangle$ oriented crystal without pre-stress should be equal to $0.75\lambda_{111}$, while it can reach $1.25\lambda_{111}$ under a compressive pre-stress. Therefore, to realize a better understanding of the enhanced magnetostriction in $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ after magnetic annealing, spontaneous magnetostriction λ_{111} was determined from the intrinsic lattice distortion reflected by high-resolution synchrotron XRD, as shown in Fig. 8. It can be seen from Fig. 8 visibly that split peaks of $\{440\}$ and $\{220\}$ reflections, where the corresponding intensity ratios are 1:1 and 1:3, respectively. Therefore, the distortion angle β , which is the deviation of the angle between neighboring edges of the distorted cube, can be obtained from:

$$\frac{d_{222} - d_{\bar{2}22}}{d_{222}} \approx \frac{4}{3} \cos \beta \quad (2)$$

where d_{222} and $d_{\bar{2}22}$ denote the crystallographic plane distances in pseudo-cubic indices (hkl). For

Fig. 8 Splitting of $\{800\}$, $\{440\}$, $\{222\}$ peaks in synchrotron XRD pattern of $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ powders

$\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$, the β is calculated to be 89.92° by using Eq. (1). In addition, the spontaneous magnetostriction M_s direction is observed to be along $[111]$, along with the rhombohedral crystal symmetry. Then, the spontaneous magnetostriction is

$$\lambda_{111} = \lambda_f - \lambda_i = \frac{a_f - a_i}{a_f} = 1.393 \times 10^{-3} \quad (3)$$

where subscripts f and i refer to the final and initial states, respectively. The data in Table 1 also shows that for the untreated $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$, λ_s under 0 MPa can only reach 1/2 of λ_{111} , which might be ascribed to the unperfected crystal orientation. After magnetic annealing, the ratio of λ_s to λ_{111} raised to 0.63. In addition, magnetostriction “jump” effect also results in a dramatic improvement of the ratio. In case of 30 MPa, λ_s/λ_{111} for the untreated $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ is enlarged to 0.86 while it further increases to 1.08 after magnetic annealing. Therefore, the magnetically annealed $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ polycrystals exhibit a remarkable magnetostrictive performance. In addition, the magnetostrictive performance might be further improved if the specimen possessed a better crystal orientation.

4. Conclusions

Transverse magnetic annealing indeed enhances the magnetostriction performance of $\langle 110 \rangle$ oriented $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ polycrystals because of the induced additional anisotropy, which can be manifested from the magnetic hysteresis loops. In comparison with the untreated specimen, although magnetic annealing results in a slight increment in hysteresis width, the corresponding value is still much lower than that of TbDyFe GMM. Especially, in combination with its spontaneous magnetostriction, the magnetically annealed $\langle 110 \rangle$ oriented polycrystals $\text{Tb}_{0.26}\text{Dy}_{0.54}\text{Ho}_{0.20}\text{Fe}_2$ exhibits a remarkable magnetostriction performance with relatively low hysteresis. The present work demonstrates that magnetic anneal-

ing is very useful to improve magnetostriction performance of rare-earth giant magnetostrictive materials even it possesses a relatively small intrinsic magnetocrystalline anisotropy constant.

Acknowledgements

This work was supported by the Foundation for the Author of National Excellent Doctoral Dissertation of China (No. 201037), the Fundamental Research Funds for the Central Universities (No. 2012QNA4007) and the Projects of Nonprofit Technology & Research in Zhejiang Province (No. 2013C31025).

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