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Magnetostrictive and Magnetic Properties of $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ Fiber/Epoxy Composites

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In this paper, we fabricated novel magnetostrictive composites by embedding <110>-oriented Tb-Dy-Ho-Fe fibers in an epoxy matrix. The magnetostrictive and magnetic properties (magnetostriction, magnetization, piezomagnetic coefficient and relative permeability) of the proposed composites were measured, analyzed, and compared to those of $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ alloy and $Tb_{0.3}Dy_{0.7}Fe_2$ continuous-fiber/epoxy composites. $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ continuous-fiber/epoxy composites have a saturation magnetostriction (λ_s) of 840 ppm and saturation magnetization (M_s) of 0.75 T. Their piezomagnetic coefficient exhibits a maximum value (8.2 μm/kA) at 19 kA/m. These proposed composites exhibit a large magnetostriction in high magnetic fields (> 400 kA/m) and a large relative permeability in low magnetic fields (< 100 kA/m). This result indicates that the given composites perform better than the $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ alloy and $Tb_{0.3}Dy_{0.7}Fe_2$ fiber/epoxy composites. Thus, the composites with characteristics of high sensitivity and large magnetostriction can be used in the field of ultrasonic sensing.

Keywords : magnetostrictive composites, Tb-Dy-Ho-Fe alloy, epoxy, fiber

1. Introduction

Tb-Dy-Fe alloy (Terfenol-D) has large magnetostriction (1000-2000 ppm) at room temperature. It has been widely applied in transducers and microdisplacement actuators [1-3]. However, the disadvantages of mechanical brittleness, low permeability and eddy current loss of Terfenol-D have limited its application in ultrasonic transducers and sensors. Therefore, the magnetostrictive composite system was put forward to overcome these shortcomings. As reported, polymer-bonded Terfenol-D composites can increase operational bandwidths and reduce mechanical brittleness and costs [4-6]. However, at the same time, the crystal structure damage and oxidation caused in the shattering process will lead to the degradation of the performance of Terfenol-D particles. The reported saturation magnetostriction loss of a magnetostrictive composite material is approximately 20 %-40 % [7, 8].

In view of this work, C. Y. Lo developed a kind of Terfenol-D continuous-fiber/epoxy composite material [9]. The Terfenol-D fibers were cut from a <112>-orientated Terfenol-D plate with a size of 1 mm × 1 mm × 4 mm. According to his method, a good balance was achieved between the low eddy current of the composites and large magnetostriction of the monolithic Terfenol-D, leading to an enhanced saturation magnetostriction reaching 80-90 % of the Terfenol-D rod [10, 11]. Terfenol-D fiber has been applied to piezoelectric/magnetostrictive composite materials and is expected to be applied in energy harvesting and sensing.

However, there are still some disadvantages, such as a high saturation magnetic field, large magnetic hysteresis and low relative permeability, of Terfenol-D composites, making it a difficult product to work with under high-frequency or low-magnetic-field conditions. Especially, the low relative permeability means the composites has low magnetization and small magnetiostriction when applied a low magnetic field. This undesirable property will reduce the sensitivity when the composites are used in sensory devices or decrease the displacement when the composites are used in actuators and transducers. It has been found that the incorporation of the element Ho can reduce the

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magnetocrystalline anisotropy of a Tb-Dy-Fe system. Therefore, some scholars began to study the Tb-Dy-Ho-Fe four-phase alloy. S. C. Busbridge and A. R. Pirecy first introduced the magneto-elastic properties and anisotropy compensation of $Tb_xDy_yHo_zFe_2$ [15]. Next, the magnetostrictive and hysteresis properties of <112>-orientated $Tb_xDy_yHo_{1-x-y}Fe_{1.95}$ were reported by M. Wun-Folge and and A. E. Clark [16]. Then, H. M. Jiang investigated the Ho-doping influence on the microstructure and magnetostriction of Tb-Dy-Fe alloy [17]. C. B. Jiang studied the magnetostriction of <110>-orientated $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_2$ under different compressive stresses [18]. B. W. Wang and co-workers researched the magnetostriction and its ratio to hysteresis for Tb-Dy-Ho-Fe alloy [19-21]. From these studies, it is reported that Tb-Dy-Ho-Fe alloy displays a large magnetostriction (> 800 ppm) and narrow hysteresis (< 6 mT).

These excellent properties of Tb-Dy-Ho-Fe alloy have piqued our interests. In previous work, we have already investigated the properties of polymer-bonded Tb-Dy-Ho-Fe particle/epoxy composites [22]. In this study, $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ fibers were utilized to fabricate the proposed magnetostrictive composites. A coupled magneto-mechanical-thermal preparation method was used to increase the saturation magnetostriction of the composites. According to the experimental results, the magnetostrictive and magnetic properties were investigated, and the mechanism of the proposed composites was analyzed. The details are given in the following sections.

2. Experiments

2.1. Fabrication

$Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ alloys were prepared from materials with the following purities: Tb, Dy, Ho (99.9 wt%) and Fe (99 wt%). Raw materials were melted in a vacuum furnace three times in the preparation of Tb-Dy-Ho-Fe ingots. Then, the ingots were cast in a water-cooled copper mold with a diameter of 60 mm. A magnetostrictive rod was directionally solidified by the zone-melting method, with a growth velocity of 240 mm/h. Finally, a <110>-orientated crystallization rod was formed.

The monolithic $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ was cut into continuous fibers with dimensions of 0.8 mm × 0.8 mm × 12 mm (See Fig. 1a) by using an electrical discharge machine. Figure 1b shows the fabricated composites, 1-3 type composites, with dimensions of 10 mm × 10 mm × 12 mm. Here, a one-component thermosetting type resin (HS-1901) was chosen as a matrix. The volume ratio of fiber to matrix was 9:1. The mixture was put into a tablet machine and pressed for 5 min with a compressive loading of 10 MPa. Then, the sample was cured at a temperature of 120 °C for 1.5 hours.

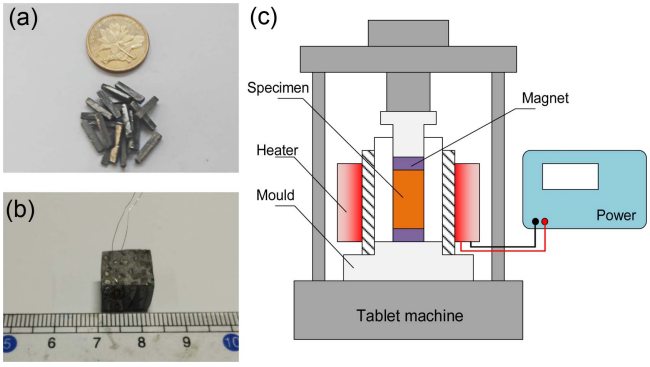

Fig. 1. (Color online) Tb-Dy-Ho-Fe composites and the fabrication equipment.

Table 1. Physical parameters of raw materials.

Material	Parameter	Value
$Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$	Saturation magnetosriction (λ_s)	810 ppm
	Young's Modulus (E)	35 GPa
	Density (ρ)	9.45 g/cm^3
$Tb_{0.3}Dy_{0.7}Fe_2$	Saturation magnetosriction (λ_s)	1250 ppm
	Young's Modulus (E)	35 GPa
	Density (ρ)	9.15 g/cm^3
Resin	Young's Modulus (E)	1.5 GPa
	Density (ρ)	1.09 g/cm^3
	Curing temperature	120 °C

To make a comparison, a sample of $Tb_{0.3}Dy_{0.7}Fe_2$ fiber/epoxy composite was also fabricated. The Terfenol-D rod was manufactured by Guansu Tianxing Rare Earth Functional Materials Co. Ltd. Table 1 shows the physical parameters of Terfenol-D, Tb-Dy-Ho-Fe alloy and epoxy resin.

2.2. Measurements

A multiparameter test system was utilized to measure the magnetostrictive and magnetic properties of the specimens. The experimental data include the magnetostriction-magnetic field (λ-H) curves, the magnetization-magnetic field (M-H) curves, the piezomagnetic coefficient-magnetic field (d_{33}-H) curves and the relative permeability-magnetic field (μ_{r33}-H) curves. The magnetostriction parallel to the longitude direction of the specimen was measured by a strain gauge with a resistance of 120 Ω. The magnetic flux density (B) was measured by a soft magnetic materials test system. The magnetization was obtained by using,

$$M = \frac{B}{\mu_0} - H \tag{1}$$

where $\mu_0 = 4\pi \times 10^{-7}$ H/m is the permeability of vacuum.

The piezomagnetic coefficient d_{33} is expressed as Eq. (2), and plotted from the λ-H curves.

$$d_{33} = \frac{\partial \lambda}{\partial H} \tag{2}$$

The relative permeability μ_{r33} is determined from,

$$\mu_{r33} = \frac{B}{\mu_0 H} \tag{3}$$

3. Results and Discussion

3.1. Magnetostriction

Figure 2 shows the λ-H curves of the $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ fiber/epoxy composites, bulk $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ and $Tb_{0.3}Dy_{0.7}Fe_2$ fiber/epoxy composites. From Fig. 2, it is found that the $Tb_{0.3}Dy_{0.7}Fe_2$ fiber/epoxy composites have the largest saturation magnetostriction (1020 ppm) at 400 kA/m. However, in low magnetic fields (< 100 kA/m), it has the smallest magnetostriction among the three samples. In contrast, the $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$ fiber/epoxy composite material displays a large magnetostriction in low magnetic fields and a low saturation magnetic field of 250 kA/m. Its saturation magnetostriction is 840 ppm, 180 ppm lower than that of the $Tb_{0.3}Dy_{0.7}Fe_2$ composites but 30 ppm higher than that of the monolithic $Tb_{0.29}Dy_{0.48}Ho_{0.23}Fe_{1.9}$.

W. Or, X. C. Guan and S. Y. Li all mentioned that the residual stresses developed in the magnetostrictive fibers during the curing process of the epoxy matrix is helpful to increasing the magnetostiction of the composites [23-25]. These built-in these built-in residual stresses can generated by heat treatment, axial compressive stress, or external magnetic field. The residual stresses effectively create a preferred non-180° domain state in the composite, which is similar to the case of applying an external pre-stress to assert an initial non-180° domain state in monolithic Tb-Dy-Ho-Fe. On the contrary, the monolithic magnetostrictive material has comparatively little initial non-180° domains since the only internal stress come from the ones generated by crystal defects and material processing. Besides this, the higher magnetostiction is also contributed to the property promotion of Tb-Dy-Ho-Fe fiber in the thermal cure process, which increases the saturation magnetostiction of magnetostrictive material.

3.2. Magnetization

The magnetization of Tb-Dy-Fe fiber/epoxy composites, monolithic Tb-Dy-Ho-Fe and Tb-Dy-Ho-Fe fiber/epoxy composites are given in Fig. 3. The saturation magnetizations of these samples are 1.1 T, 0.75 T and 0.7 T, respectively. There is only a 7 % saturation magnetization loss of the Tb-Dy-Ho-Fe fiber/epoxy composite compared to that of the Tb-Dy-Fe fiber/epoxy composites. Therefore, the Tb-Dy-Fe composites are saturated at 400 kA/m. In addition, the Tb-Dy-Ho-Fe composites reach saturation magnetization at 250 kA/m, which is very close to the saturation magnetic field of Tb-Dy-Ho-Fe alloy (220 kA/m).

3.3. Relative Permeability

Figure 4 shows the relative permeability of the three samples. With the definition of Eq. (3), the μ_{r33}-H curves are plotted from the B-H curves.

From Fig. 4, it is observed that in low magnetic fields

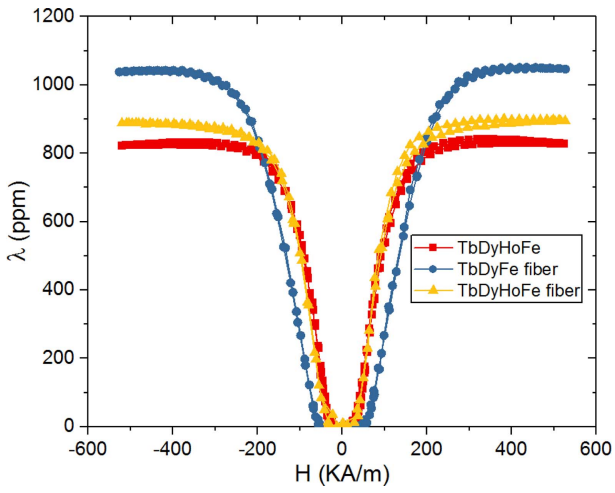

Fig. 2. (Color online) Magnetsotriction of Tb-Dy-Ho-Fe, Tb-Dy-Ho-Fe composites and Tb-Dy-Fe composites.

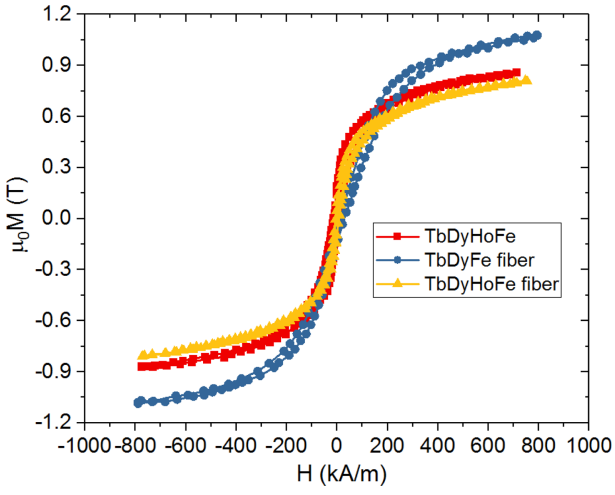

Fig. 3. (Color online) Magnetization of Tb-Dy-Ho-Fe, Tb-Dy-Ho-Fe composites and Tb-Dy-Fe composites.

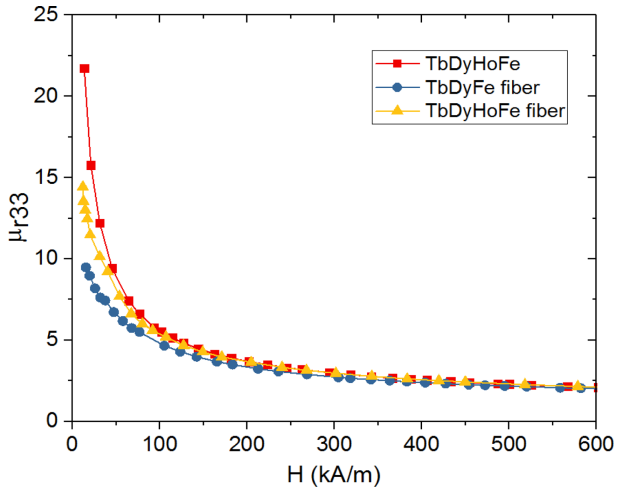

Fig. 4. (Color online) Relative permeability of Tb-Dy-Ho-Fe, Tb-Dy-Ho-Fe composites and Tb-Dy-Fe composites.

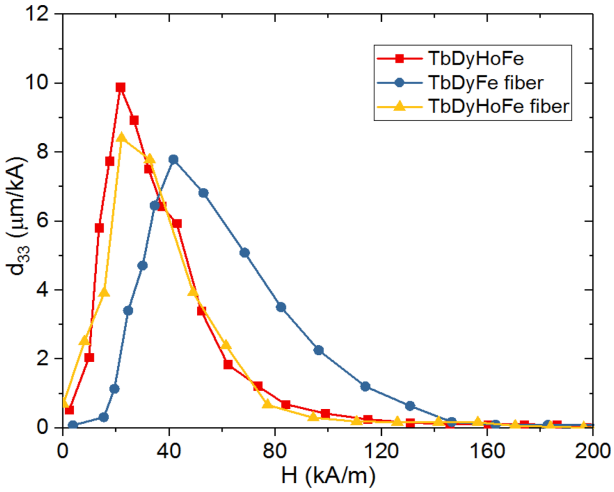

Fig. 5. (Color online) Piezomagnetic coefficient of of Tb-Dy-Ho-Fe, Tb-Dy-Ho-Fe composites and Tb-Dy-Fe composites.

(< 100 kA/m), the value of relative permeability of the Tb-Dy-Ho-Fe fiber/composites is between those of the Tb-Dy-Ho-Fe alloy and Tb-Dy-Fe fiber composites. At 50 kA/m, their relative permeabilities are 6.7, 9.1 and 9.6, respectively. The relative permeability of the Tb-Dy-Ho-Fe composite is 7 % lower than that of the Tb-Dy-Ho-Fe alloy and 35 % higher than that of the Tb-Dy-Fe fiber composites. The loss is due to the demagnetization effect in the composite and the non-180° domain state induced by the residual stress. The higher relative permeability of the proposed composites compared to the Tb-Dy-Fe fiber composites means that it is more sensitive in low magnetic fields.

3.4. Piezomagnetic Coefficient

The value of the piezomagnetic coefficient for the composites can be obtained by using plots similar to those shown in Fig. 2. This value is obtained by fitting a 6th-order polynomial to the loading portion of the curve. The pointwise slope of the polynomial corresponds to d_{33} as a function of the magnetic field (see Fig. 5). The initial increase is caused by the increase in the non-180° domain-wall motion, and d_{33} reaches its maximum value when the non-180° domain-wall is maximized. d_{33} decreases with respect to an increasing magnetic field due to the domain saturation.

From Fig. 5, it was found that for the Tb-Dy-Fe composites, the magnitude of d_{33} increases rapidly from 0-40 kA/m and displays a broad maximum (7.6 μm/kA) at 50 kA/m; the monolithic Tb-Dy-Ho-Fe has the largest d_{33} at 16 kA/m and the largest change rate in the magnetic field range of 0-16 kA/m, and its maximum value is 9.8 μm/kA; d_{33} of the proposed composite material reaches its peak (the maximum value is 8.2 μm/kA) at 19 kA/m.

4. Conclusion

In this study, new magnetostrictive composites with epoxy resin-coated magnetostrictive fiber were fabricated. To obtain a large magnetostriction in low magnetic fields, Tb-Dy-Ho-Fe fiber was chosen instead of the Tb-Dy-Fe fiber used in the composites. Furthermore, residual stress was introduced during the curing process to improve the saturation magnetostriction of the proposed composites. With this method, large magnetostrictions were obtained in both low and high magnetic fields. The properties of the composites were tested and analyzed, and the following conclusions were drawn:

(i) For the proposed composites, the replacement of Tb-Dy-Fe fiber with Tb-Dy-Ho-Fe fiber is performed to obtain a large magnetostriction from Tb-Dy-Ho-Fe in low magnetic fields. Thus, the narrow magnetic hysteresis of the alloy is inherited in the presented composites.

(ii) The enhancement of the saturation magnetostriction is caused by the residual stress derived during the preparation process and the stress concentration of the inclusion in the matrix under external compression. Notably, this method will cause an increase of the saturation magnetic field for magnetostrictive composites with built-in residual stress.

(iii) Through the optimization, we got a composite material with better comprehensive performances. The saturation magnetostriction of the proposed composites is 840 ppm, with a low saturation magnetic field as 250 kA/m. And the maximum piezomagnetic coefficient is 8.2 μm/kA at 19 kA/m.

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References

[1] S. Karunanidhi and M. Singaperumal, Sens. Actuators A **157**, 185 (2010).
[2] W. Kim and A. Sadighi, IEEE Trans. Mechatronics **15**, 299 (2010).
[3] R. Zhao, B. W. Wang, Q. G. Lu, L. Y. Zhang, and Q. H. Cao, J. Comput. Theor. Nanos. **13**, 7926 (2016).
[4] D. Kendall and A. R. Piercy, J. Appl. Phys. **73**, 6174 (1993).
[5] S. H. Lim, S. R. Kim, S. Y. Kang, J. K. Park, J. T. Nam, and D. Son, J. Magn. Magn. Mater. **191**, 113 (1999).
[6] C. Rodriguez, A. Barrio, I. Orue, J. L. Vilas, L. M. Leon, J. M. Barandiaran, and M. L. Ruiz, Sens. Actuators A **142**, 538 (2008).
[7] N. Nersessian, S. W. Or, and G. P. Carman, J. Magn. Magn. Mater. **263**, 101 (2002).
[8] J. Kaleta, D. Lewandowski, R. Mech, and P. Gąsior, Solid State Phenom. **154**, 35 (2009).
[9] Ch. Y. Lo, S. W. Or, and H. L.W. Chan, IEEE Trans. Magn. **42**, 3111 (2006).
[10] G. P. McKnight and G. P. Carman, Mater. Trans. **43**, 1008 (2002).
[11] S. W. Or, N. Nersessian, G. P. McKnight, and G. P. Carman, J. Appl. Phys. **93**, 8510 (2003).
[12] C. Y. Lo, S. H. Choy, S. W. Or, and H. L. W. Chan, J. Appl. Phys. **107**, 093907 (2010).
[13] K. H. Lam, C. Y. Lo, and H. L. W. Chan, J. Mater. Sci. **47**, 2910 (2012).
[14] C. M. Leung and S. W. Or, J. Appl. Phys. **117**, 17D721 (2015).
[15] S. C. Busbridge and A. R. Piercy, IEEE Trans. Magn. **31**, 4044 (1995).
[16] M. Wun-Fogle, J. B. Restorff, and A. E. Clark, J. Appl. Phys. **85**, 6253 (1999).
[17] M. H. Jiang, Z. F. Gu, G. Cheng, and X. Y. Liu, J. Rare Earths **27**, 150 (2009).
[18] H. B. Zhang, C. B. Jiang, Z. B. Wang, and H. B. Xu, J. Alloys Compd. **475**, 35 (2009).
[19] B. W. Wang, Y. Lv, G. Li, W. M. Huang, Y. Sun, and B. Z. Cui, J. Appl. Phys. **115**, 17A902 (2014).
[20] B. W. Wang, W. M. Huang, L. Weng, Y. Sun, S. Y. Cao, and Z. H. Wang, J. Rare Earths **33**, 1170 (2015).
[21] B. W. Wang, Y. Lv, G. Li, W. M. Huang, L. Weng, and B. Z. Cui, J. Appl. Phys. **117**, 17A912 (2015).
[22] R. Zhao, B. W. Wang, S. Y. Cao, and J. Xiao, Chem. Eng. Trans. **55**, 301 (2016).
[23] S. W. Or and G. P. Carman, IEEE Trans. Magn. **41**, 2790 (2005).
[24] X. C. Guan, X. F. Dong, and J. P. Ou, Acta Materiae Compositae Sinica **25**, 11 (2008).
[25] S. Y. Li, B. W. Wang, L. Weng, Y. Zhou, Y. Sun, and W. M. Huang, J. Magn. Magn. Mater. **320**, 806 (2008).