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Effect of Ho Doping and Annealing on Magnetostrictive Properties of Tb-Dy-Ho-Fe/Epoxy Composites

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The effect of Ho-doping and annealing on magnetostriction and magnetostriction hysteresis of Tb-Dy-Ho-Fe/epoxy composites was investigated in this paper. The polymer bonded $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$ composites (x=0.21, 0.25 and 0.31) were prepared and annealed in a vacuum magnetic heat treatment furnace at 20, 80, 100 and 150 °C. The microstructure of the sample was then observed by optical microscopy. The magnetostricion of the composites was studied by a standard strain gauge technique. The research results showed that when the Ho content x=0.31, the composites exhibited large magnetostriction and small magnetostriction hysteresis. When annealing temperature is 100°C, the composites have a maximum magnetostriction at 695 ppm.

1. Introduction

In recent decades, giant magnetostrictive materials (Tb-Dy-Fe alloys) have attracted great attention due to their large magnetostriction (1500-2000 ppm) at room temperature (Claeyssen et al., 1999 and Squire, 2011). Compared to other smart materials, such as shape memory alloy and piezoelectric, it exhibits obvious advantages of applications in transducers and actuators (Shigenao et al., 2001, VANNESSA and CERROLAZA, 2013, Rajan et al., 2015, Morega et al., 2016). However, the high saturation magnetic field and large magnetostriction hysteresis has restricted its applications (Zhao et al., 1998). The investigations of magnetostrictive hysteresis have shown that the addition of Ho element to Tb-Dy-Fe alloys can reduce the hysteresis width and improve the magnetostriction in the low magnetic field (Busbridge and Piercy, 1999, Wang et al., 2014, 2015, 2015 and 2016).

The excellent magnetostrictive properties of Tb-Dy-Ho-Fe alloys can be utilized with polymer bonding technology (Lim et al., 1999, Jonhson and Amirouche, 2008, Dong et al., 2011) to fabricate the Tb-Dy-Ho-Fe/epoxy composites. This composites exhibits advantages of superior mechanical properties, high frequency performance and low cost. Thus, Tb-Dy-Ho-Fe/epoxy composites is hoped to find applications in high sensitivity magnetometers and ultrasonic transducers (Karafi et al., 2003, Wang and Feng, 2013). In summery, the research on the magnetostrictive properties of Tb-Dy-Ho-Fe/epoxy composites is of great significance.

In this study, the effect of the Ho-doping and annealing temperature on the magnetostriction and magnetostriction hysteresis of Tb-Dy-Ho-Fe/epoxy composites was studied. And we fabricated the $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$/epoxy composites with different Ho content, and annealed the composites at different temperatures. Then we studied the magnetostriction and magnetostriction hysteresis, and the influence of annealing temperature on the magnetostriction of the composites.

2. Experiments

The magnetostrictive compounds were prepared from materials of the following purity: Tb, Dy, and Ho (99.9 wt.%), Fe (99.8 wt.%). The $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$ (x=0.21, 0.25 and 0.31) compounds were

melted in an arc furnace under high purity argon. The as-cast samples wrapped in Mo foil were sealed in a silica tube filled with high purity argon and were homogenized at 1000 °C for 24 h and at 950 °C for 120 h. The Tb-Dy-Ho-Fe alloys was crushed in absolute ethanol by a ball milling machine, into the particle size range of 75-180 μm.
Double component epoxy resin (AB resin) was used as the matrix. Its working temperature is 200 °C. The mass fraction of A-component, B-component and magnetostrictive particles is 5:1: 100. Mixing process of the resin and magnetostrictive particles was operated in the glove box. Then the mixture was transferred to the mould, where it was compressed by the press machine at 2.5 MPa for 5 min. Finally, the mixture was cured at room temperature, made into 10×10×5 mm samples. This method was used to prepare the samples with Ho content of x=0.21, 0.31 and 0.25.
The microstructure of the composites was observed using the optical microscope at first. And then the magnetostriction and the magnetostriction hysteresis of the samples were measured by the magnetic measurement system. The samples were annealed in a vacuum magnetic field heat treatment furnace, and the heating temperatures were 25, 80, 100 and 150 °C. At last, the annealed of the samples were tested and their magnetostriction curves were compared.

3. Results and Discussion

3.1 Microstructure

The optical microscope was utilized to observe the microstructure of the composites. Figure 1 shows the microsructure of $Tb_{0.25}Dy_{0.45}Ho_{0.31}Fe_{1.9}$/epoxy composites.From the figure, we can see that the black part is the epoxy matrix, the white area is the magnetostrictive particle. The particles are uniformly distributed in the matrix, and the magnetostrictive particles are aligned in the direction of the long axis of the composites. The proposed composites has typical pseudo 1-3 structure.

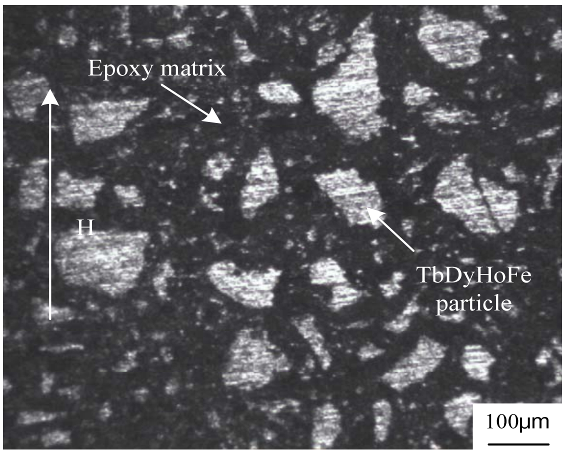

Figure 1: Optical micrograph of the $Tb_{0.25}Dy_{0.45}Ho_{0.31}Fe_{1.9}$/epoxy composites

3.2 The effect of Ho content

The influence of Ho element on the magnetostriction of Tb-Dy-Ho-Fe alloy has been researched in literature (JIANG et al., 2009, Wang et al., 2014) already. Therefore, it is reasonable to suppose that magnetocrystalline anisotropy constant K_1 and magnetostrictive hysteresis decreases with increasing Ho content in range of $0.1<x<0.6$. In this section, the Tb-Dy-Ho-Fe/epoxy composites with different Ho content are tested to investigate whether this rule applicable to the magnetostrictive composites.
Figure 2 exhibits the λ-H curves with varying Ho content. When x=0.21, the saturation magnetostriction λ_s is 480 ppm. And the magnetostriction λ is 82 ppm at 100 kA/m, λ=220 ppm at 200 kA/m. Its saturation magnetic field is 760 kA/m. When x=0.25, the composite is saturated at 755 kA/m and λ_s is 514 ppm. In the meantime, its magnetostriction is 110 ppm at 100 kA/m, and λ=263 ppm at 200 kA/m. When x=0.31, the saturation magnetostriction and saturation magnetic field of the sample are 605 ppm and 750 kA/m, respectively. At 100 kA/m applied magnetic field, its magnetostriction increase to 202 ppm, and λ=387 ppm when H is 200 kA/m. From Figure 2 we found an obvious growth that with the increasing of Ho content, λ_s of the composites is also increased, and the saturation magnetic field strength is decreased. In the low magnetic field (<100 kA/m), the composites' magnetostriction is greatly improved when x=0.31.

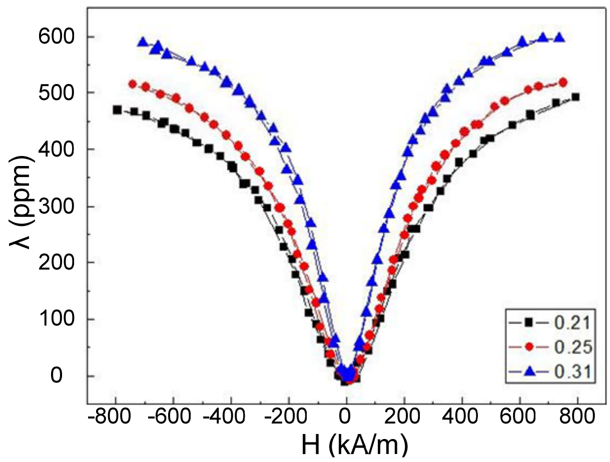

Figure 2: The λ-H curves of $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$/epoxy composites with x=0.21, 0.25 and 0.31

The magnetostriction hysteresis width of $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$/epoxy composites was tested at 50, 100, 150 and 200 kA/m applied magnetic field, respectively. Its hysteresis characteristics are shown in Figure 2 and Figure 3. Figure 3 shows the variation of hysteresis width W_h with Ho content. When x=0.21, the maximum hysteresis width is 9.1 kA/m at 100 kA/m applied magnetic field. The maximum hysteresis width appears at 100 kA/m and its value is 7 kA/m when x=0.25. And when x=0.31, the maximum hysteresis width is 3 kA/m at 150 kA/m. These events that the composites has the minimum hysteresis width when the Ho content is 0.31. The improvement in the hysteresis width by Ho element is particularly evident in the low magnetic field.

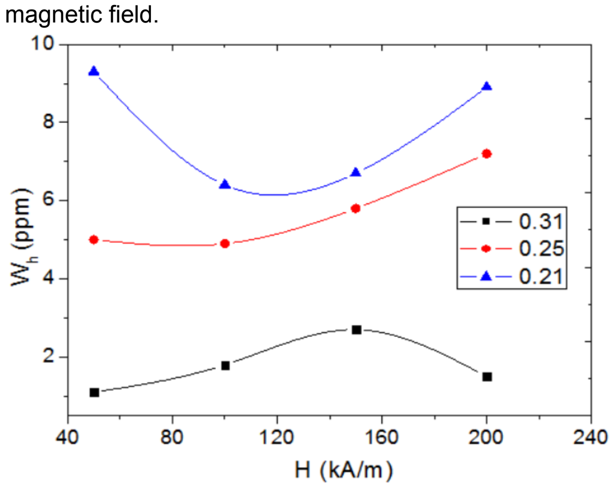

Figure 3: The hysteresis width of $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x$+$(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$/epoxy composites with x=0.21, 0.25 and 0.31

In Figure 4, the ratio of magnetostriction to hysteresis width (λ/W_h) at megnetic field of 50, 100, 150, 200 kA/m are shown, which is determined by the measured magnetostriction and hysteresis width curves shown in Figure 2 and Figure 3. From Figure 4 it can be seen that the ratio rises with increasing x, and it shows a peak at 200 kA/m applied magnetic field. And this events that the $Tb_{0.285}Dy_{0.63}Ho_{0.085}Fe_{1.9}$/epoxy composites performs both large magnetostriction and narrow magnetostriction hysteresis.

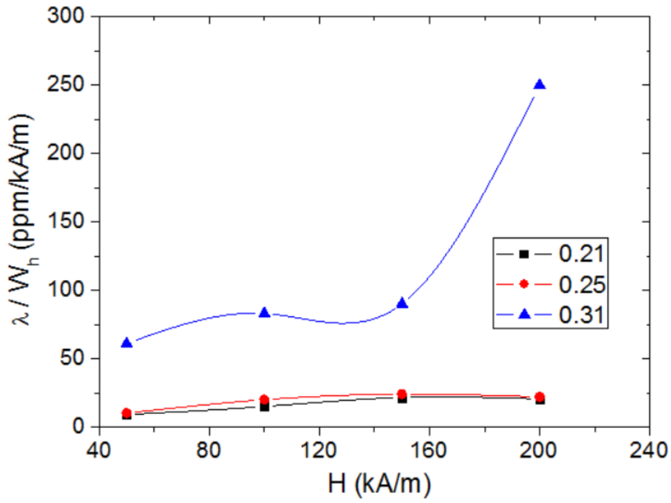

Figure 4: The ratio of magnetostriction to hysteresis width (λ/Wh) of $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x+(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$ /epoxy composites with x=0.21, 0.25 and 0.31

3.2 The effect of annealing

In order to investigate the influence of the annealing effect on the magnetostrictive performance, the magnetic field heat treatment experiments were performed on the composites. The annealing temperatures chosen were 20, 80, 100 and 150 °C.

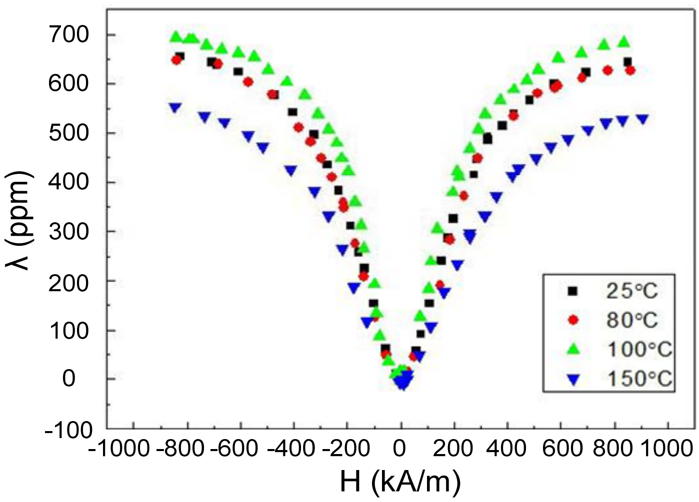

Figure 5: Magnetostriction curves after annealing at 25, 80, 100 and 150 °C (x=0.31)

As annealed at 25 and 80 °C, the magnetostriction didn't show obviously change. When the heating treatment temperature was 100 °C, the saturation magnetostriction reached 695 ppm. However, when the temperature was increased further to 150 °C, the magnetostrictive properties of the sample were greatly decreased, and the value was 550 ppm. As discussed in references (Or et al., 2003, Na et al., 2004 and Dong et al., 2010), there are several factors determining the magnetostrictive performance of the composites: the shape, size and magnetostriction of magnetostrictive particles, the young's modulus of the composites. In this The optimum annealing temperature has been found at 100 °C. At this temperature, the rich rare-earth phase (Tb, Dy, Ho) in Tb-Dy-Ho-Fe alloys will be reduced. Due to this, the magnetostriction performance will be improved. Also noted is that the residual thermal stress generated during the heat process will increase the saturation magnetostriction of the composites. Conversely, over-heating will reduce the performance of Tb-Dy-Ho-Fe particles in the composites, as well as the young's modulus of the composites.

4. Conclusions

In this work, the influence of Ho content and annealing on the magnetostriction performance of the $(Tb_{0.15}Ho_{0.85}Fe_{1.9})_x+(Tb_{0.3}Dy_{0.7}Fe_{1.9})_{1-x}$/epoxy composites was studied. In order to find the optimum Ho content and heat treatment temperature, we fabricated the samples with Ho content at x=0.21, 0.25 and 0.31. And annealed these samples by a magnetic vacuum magnetic field heat treatment furnace with heating temperatures of 25, 80, 100 and 150 °C. We also experimented with the magnetostrictive performance, and found some patterns as follows:

(1) The performances of magnetostrictive composites will be improved by Ho element. With increasing of *x*, the composites' magnetostrictive hysteresis was reduced and the saturation magnetostirction was promoted. When x=0.31, a large magnetostriction (605 ppm) and narrow magnetostrictive hysteresis (W_h is 3 KA/m at 150 KA/m applied magnetic field) were observed. Which illustrated that Ho-doping is beneficial to increasing the magnetostriction under low magnetic field and reducing the hysteresis width of Tb-Dy-Ho-Fe/epoxy composites.

(2) An appropriate heat treatment temperature is helpful for the properties of magnetostrictive composites. When annealed at 100 °C, the saturation magnetostriction is increased to 695 ppm, and the magnetostriction property at low magnetic field is retained. However, it is noted that the over-heating in the heat treatment process will cause damages and decline the magnetostrictive performance of the Tb-Dy-Ho-Fe/epoxy composites.

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