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Magnetostriction of grain-aligned $Tb_{0.3}Dy_{0.7}Fe_{1.95}$

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Abstract

Commercial purity raw materials have been used to prepare grain-aligned $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ rods. The effects of material structure and heat treatment on magnetostriction were investigated. It was found that grain orientation was crucial to the magnetostrictive properties and had a stronger effect than lamellar spacing. The effect of heat treatment depended greatly on material structure and annealing temperature. High temperature annealing up to 1150 °C was needed for samples with narrow lamellar spacing to get an obvious rise in their compressive magnetostrictive properties. Under 10 MPa compressive stress, typical magnetostriction values were 1500 ppm in 4 kOe applied field and 1000 ppm in 1.5 kOe. The magnetostriction variation for one lot of 10 rods was no more than 200 ppm.

Keywords: Magnetostriction; Directional solidification; Rare earths; Low purity

1. Introduction

The giant magnetostrictive alloy $Tb_{0.3}Dy_{0.7}Fe_{\sim 1.95}$ (Terfenol-D) possesses huge linear magnetostriction (λ) and low magnetocrystalline anisotropy at room temperature [1]. Its huge magnetostriction arises solely from a large λ_{111} of the $REFe_2$ Laves intermetallic phase when $\lambda_{100} \ll \lambda_{111}$. The RE-rich (RE, rare earth) composition provides a ductile network of grain boundary RE-rich phase which enhances the mechanical properties. Previous studies have established that although ⟨111⟩-aligned single crystals are very difficult to grow, it is fairly easy to obtain ⟨112⟩-twinned single crystals and ⟨112⟩ grain-aligned rods [2,3]. However, those studies were mainly on expensive raw materials of high purity (99.99% or 99.999%) and less has been reported about the relationship between material structure and magnetostrictive properties, especially compressive magnetostriction. Recently, interest in this alloy has become more application oriented [4,5]. To reduce its cost, it is necessary to study the effects of impurities, i.e. to determine the effects of different structure factors and the property level of the low purity alloy.

In this paper, commercial purity (99.9%) raw materials were used to prepare $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ grain-aligned rods. The effects of material structure (grain alignment, lamellar spacing) and heat treatment on magnetostriction were investigated. Typical magnetostriction values were 1500 ppm in 4 kOe applied magnetic field and 1000 ppm in 1.5 kOe under a compressive stress of 10 MPa. The magnetostriction variation for one lot of 10 rods was no more than 200 ppm.

2. Experimental

Raw materials were of 99.9 wt.% purity. Master alloy rods (diameter 8 mm) were prepared in an r.f. induction casting furnace using alumina crucibles. Grain-aligned rods were made by zone melting in alumina tubes (inner diameter 8 mm) at 0.5–30 mm min^{-1} growth rates with a very high temperature gradient. The rods were then annealed at 950–1150 °C for 1–2 h in an argon atmosphere and air cooled.

Magnetostriction measurements were made on samples 40 mm long using standard strain gauge techniques. A gas pressure cell was employed to produce 0–10 MPa axial prestress. $(d\lambda^p/dH)_{max}$ was calculated from the λ–H curve. Chemical analyses showed an alloy composition near $Tb_{0.3}Dy_{0.7}Fe_{1.95}$, while O and N contents were 800 and 70 ppm respectively. Microstructure observations revealed that each sample has at least three grains. Small amounts of second phase and RE oxides are present in all samples. RE oxides always exist as inclusions inside the grains and they agglomerate. The RE-rich phase is distributed lamellarly parallel to

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the rod axis. The average lamellar spacings were determined. Thin sections were cut from the ends of the rods and analysed with X-ray techniques to determine axial grain orientation. Solidification studies will be discussed in detail elsewhere.

3. Results and discussion

3.1. Typical magnetostrictive properties

Fig. 1 illustrates a typical plot of the magnetostrictive properties of low purity grain-aligned $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ rods. Besides the 1100 ppm saturation magnetostriction there is an obvious "stress effect". Magnetostrictive behaviour is greatly improved by the prestress. Under a 4 MPa compressive stress the compressive magnetostriction λ^p reaches 1400 ppm in 3 kOe applied field and $(d\lambda^p/dH)_{max}$ is 1.5 ppm Oe^{-1}, which is twice the value of 0 MPa. At 10 MPa, λ^p has a value of 1000 ppm in 1.5 kOe, increasing to 1600 ppm in 5 kOe. Such a property level can satisfy many practical demands, especially when coupled with a low material cost.

From Fig. 1 it can also be seen that λ^p has a negative initial magnetostriction of −25 ppm at 10 MPa. This phenomenon is regarded as one feature of high quality samples [5]. A maximum of −120 ppm at about 2 MPa and a decrease to −20 ppm at about 6 MPa have been reported [6].

Table 1 compares the present results with previous results of Verhoeven et al. [7]. The property differences are mainly with $(d\lambda^p/dH)_{max}$ and $\lambda^p_{2.5\ kOe}$. If the differences are attributed to only the approximately 1000 ppm content of non-metallic elements, their harmful effects can be estimated very roughly as −20 ppm for $\lambda^p_{2.5\ kOe}$ and −0.1 ppm Oe^{-1} for $(d\lambda^p/dH)_{max}$ per 100 ppm of impurities. Assuming that the RE oxide is RE_2O_3, the RE oxide content can be calculated as less than 1 wt.% from the known 800 ppm oxygen content. This accounts for the small effect on λ_s and the obvious decrease in the structure-sensitive properties $(d\lambda^p/dH)_{max}$ and $\lambda^p_{2.5\ kOe}$, because the 1 wt.% of impurities will impede the magnetization process greatly by pinning domain walls and hindering the rotation of domain moments [8]. It is very fortunate that the impurity inclusions tend to congregate, doing far less harm than if distributed widely.

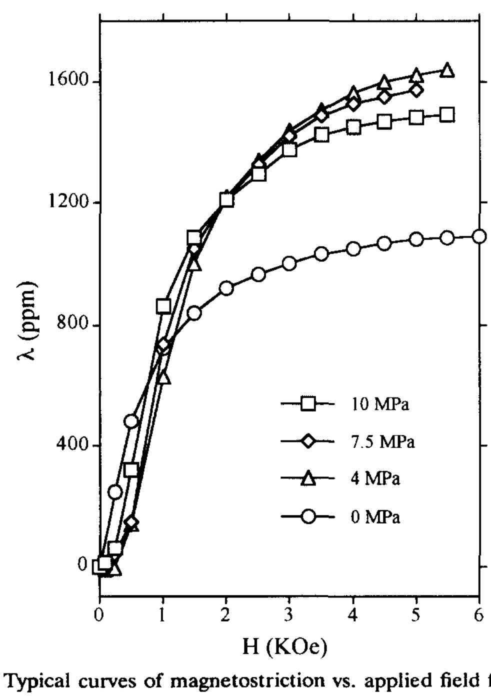

Fig. 1. Typical curves of magnetostriction vs. applied field for grain-aligned rods under various compressive stresses.

Table 1
Comparison with previous results

Sample	λ^p (ppm) (nearly saturated)	$\lambda^p_{2.5\ kOe}$ (ppm)	$(d\lambda^p/dH)_{max}$ (ppm Oe^{-1})
Present (p=7.5 MPa)	1580	1300	1.8
MB samples [a] [6]	1600	1500	2.7
FSZM samples [a] [6]	1800	1650	4.0

[a] The data are the average values of the reported data measured under 6.9 MPa; the FSZM samples are those with at least three grains.

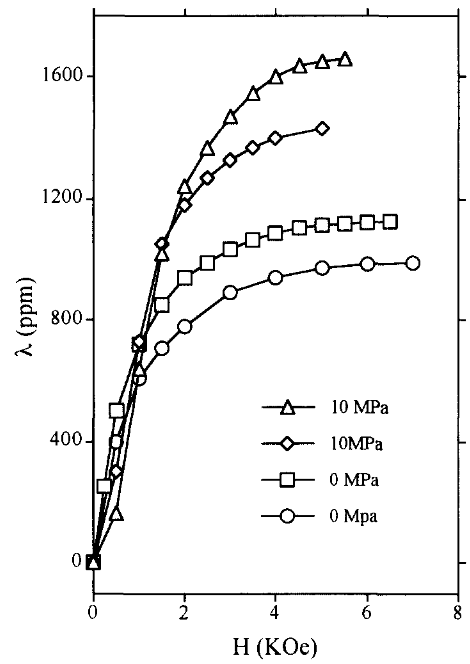

Fig. 2. Magnetostriction range of one lot of 10 rods with different strain gauge locations on four rods.

Fig. 2 shows measurements on one lot of 10 rods with different strain gauge location on four rods. Their differences are no more than 200 ppm, with the highest value being 1600 ppm in 4 kOe at 10 MPa (the highest bias level of the present measurement system for samples of 8 mm diameter). This variation is related to the grain size and orientation at the strain gauge location.

3.2. Effects of grain orientation and lamcllar spacing

Table 2 summarizes the effects of grain orientation and lamellar spacing. The value of λ_s is only dependent on the former, while the compressive magnetostriction is sensitive to both. Grain orientation affects the properties more strongly than lamellar spacing, especially $(d\lambda^p/dH)_{max}$. This can be attributed to the effect of inhomogeneous internal stresses caused by grain disorientation during the magnetization process. The low values for narrowly spaced samples are due to the unfavourable effect of the narrowly distributed RE-rich phase on the magnetization process.

The compressive magnetostriction curves of variously spaced samples are shown in Fig. 3. Comparison reveals that (1) the magnetostriction difference is bigger between the 80 and 40 μm samples than between the 150 and 80 μm samples and (2) high prestress reduces the magnetostriction difference between the various curves. These results indicate that the effects of the RE-rich phase are more obvious in narrowly spaced samples or under low prestress. One important reason for this is the effect of internal stresses produced during the magnetization process. The internal stress occurs because of the magnetostriction difference between the $REFe_2$ phase and the RE-rich phase at room temperature. It is very detrimental to magnetization because of the huge magnetostriction of the $REFe_2$ phase. In narrowly spaced samples the induced stresses are higher and will become more and more severe with narrowing of the spacing. This leads to the more harmful effect of the RE-rich phase in the narrowly spaced case.

Axial prestress favours the preferential perpendicular orientation of the moments and therefore leads to a larger domain moment rotation. Low prestress is sufficient for widely spaced samples to reorient most of their moments, while higher prestress is needed for narrowly spaced samples to overcome the far greater internal stresses which inhibit moment reorientation.

Table 2
Magnetostrictive properties of grain-aligned $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ samples (p = 10 MPa)

Degree of axial ⟨112⟩ orientation	Lamellar spacing (μm)	λ_s (ppm)	λ^p (ppm) (nearly saturated)	$\lambda^p_{2.5\ kOe}$ (ppm)	$(d\lambda^p/dH)_{max}$ (ppm Oe^{-1})
Strong	150	1100	1620	1340	1.8
Middle	150	1000	1230	1075	0.75
Weak	150	900	920	800	0.4
Strong	150	1100	1620	1340	1.8
Strong	80	1100	1550	1227	1.2
Strong	40	1100	1300	980	0.7

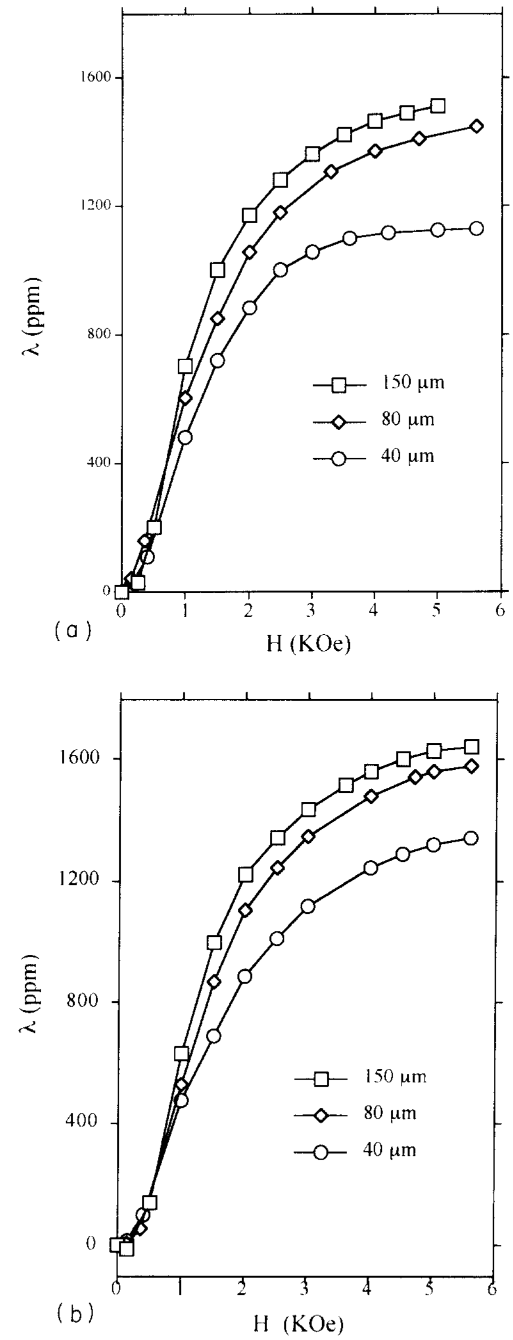

Fig. 3. Compressive magnetostriction curves of samples with various spacings at (a) 4 MPa and (b) 10 MPa.

Although narrow spacing is not good for magnetostrictive properties, it helps to improve the strength and toughness of the materials because of the dense interconnected skeleton network of the ductile RE-rich phase [9]. Mechanical compression tests show that the compressive fracture strength increases from 400 to 760 MPa as the spacing decreases from 150 to 40 μm. Considering the smaller magnetostriction decrease in samples with spacing larger than 80 μm, it is better to choose such ductile materials from the standpoint of applications.

3.3. Effect of heat treatment

The effectiveness of heat treatment can be seen in Fig. 4. Although the effect of heat treatment on λ_s is not obvious, it improves the compressive magnetostriction greatly. The value of λ^p reaches 1500 ppm in 4 kOe field under 10 MPa, while $(d\lambda^p/dH)_{max}$ increases from 0.6 to 2.0 ppm Oe^{-1}. For this high quality sample (strong ⟨112⟩ orientation, 150 μm spacing), 950 °C/1 h heat treatment is enough to achieve the largest increase, in agreement with the results of Verhoeven et al. [7]. However, for samples with narrow lamellar spacing, which always have little or no "stress effect" before annealing, a higher annealing temperature is needed to get an obvious increase. As shown in Fig. 5, for the sample with 40 μm spacing, 950 °C/1 h and 1050 °C/1 h conditions cannot produce an obvious effect; the effect becomes apparent only after annealing at 1150 °C for 1 h. However, for samples with bad grain alignment and narrow spacing, even the 1150 °C/24 h annealing is ineffective, just as for randomly oriented samples.

Verhoeven et al. have proposed that the reason for this improvement is that the heat treatment changes the residual stress distribution of the as-solidified materials owing to localized melting around the RE-rich phase and thereby either reduces the pinning of domain walls or enhances the rotation of domain moments [7]. The melting occurs in the temperature range near the eutectic reaction, which was measured as occurring at 890 °C [10]. Their results showed that the 950 °C/1 h annealing is sufficient for the twinned single crystals and grain-aligned rods made from high purity raw materials. In the present experiments the high temperature annealing is effective for most of the grain-aligned samples, while the 950 °C/1 h annealing is sufficient only for some samples, always the high quality samples. Thus 1150 °C/2 h annealing is adopted as a routine process applied to all as-solidified rods. The reason for the necessity of a high temperature annealing may lie in the more severe residual stresses caused by the narrow spacing and the high cooling rate of the high temperature gradient furnace. The detailed mechanism may involve a change in the boundary phase and stress relief during annealing.

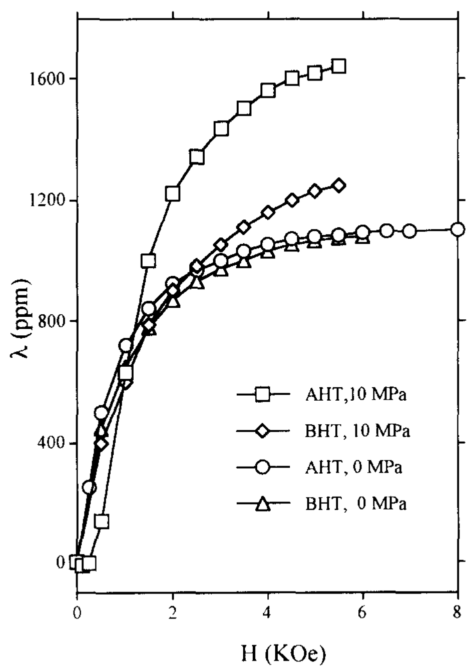

Fig. 4. Magnetostriction curves before (BHT) and after (AHT) heat treatment (150 μm).

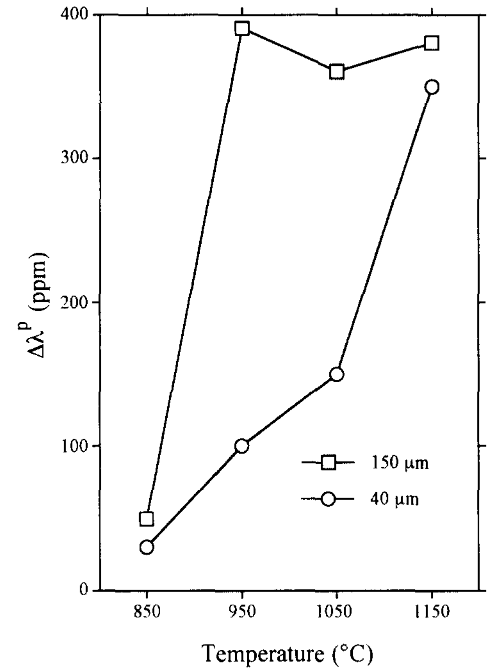

Fig. 5. Variation in $\Delta\lambda^p$ as the annealing temperature increases. $\Delta\lambda^p$ is the increase in λ^p after heat treatment.

4. Conclusions

(1) Typical magnetostrictive properties of low purity grain-aligned $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ were found to be 1500 ppm in 4 kOe applied field and 1000 ppm in 1.5 kOe under 10 MPa compressive stress. This material should be able to meet a lot of application demands, although its $(d\lambda^p/dH)_{max}$ value is less than 2.0 ppm Oe^{-1}, which is lower than the 2.7 ppm Oe^{-1} for high purity materials.

(2) Grain orientation affected magnetostrictive properties more strongly than lamellar spacing, especially $(d\lambda^p/dH)_{max}$. The effects of lamellar spacing were obvious in more narrowly spaced samples, but were reduced under higher prestress.

(3) High temperature annealing up to 1150 °C was needed for samples with narrow lamellar spacing to get an obvious "stress effect".

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