Effect of texture on the magnetostriction of grain-aligned composite Terfenol-D

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Grain-aligned composite Terfenol-D was fabricated via unidirectional solidification, followed by the infiltration of epoxy. The fraction of the $R\text{Fe}_2$ phase in composite Terfenol-D depends on the iron/rare earth (RE=Tb, Dy) ratio of the as-grown Terfenol-D and the direction of the $R\text{Fe}_2$ phase in the as-grown Terfenol-D changes from $\langle 110 \rangle$ to $\langle 111 \rangle$ with an increasing iron/rare earth ratio. For a series of grain-aligned composite Terfenol-D with different $R\text{Fe}_2$ fractions, the magnetostriction was found to increase from 813 to 1013 ppm with increases in the $R\text{Fe}_2$ fraction. A model for the magnetostriction of grain-aligned composite Terfenol-D was developed based on the modified uniform strain condition. The modified uniform strain model was then used to predict the magnetostriction of grain-aligned composite Terfenol-D from the fraction of the $R\text{Fe}_2$ phase, the elastic modulus for each crystal direction of the $R\text{Fe}_2$ phase obtained in a texture analysis, and the saturation magnetostriction value of each preferred direction. The predicted magnetostriction of grain-aligned composite Terfenol-D, which considers the elastic modulus with respect to the crystal direction of the magnetostrictive phase, is in agreement with the experimental results. In conclusion, in grain-aligned composite Terfenol-D fabricated using the crystal growth technique the preferred direction should be considered for predicting the magnetostriction of grain-aligned composite Terfenol-D. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2402347]

I. INTRODUCTION

Magnetostrictive composites have interesting potential applications, such as high-frequency transducers or sensors, because they can reduce the eddy current loss and improve the fracture toughness compared to monolithic Terfenol-D. In general, magnetostrictive [1-3] composites can be fabricated by integrating magnetostrictive powders embedded in a nonmetallic matrix, such as epoxy or metallic matrix (Al, Cu, or Fe); on applying a magnetic field along the longitudinal direction of the composite, this results in an insulating layer between the particles.[1,2] Although these [1-3] composites are based on magnetically chained particles producing a strain, they cannot avoid the demagnetizing effect and have much lower saturation magnetostriction than monolithic magnetostrictive materials.[3–5]

Previously, we reported the magnetostriction properties of the grain-aligned composite Terfenol-D, which consists of a unidirectionally solidified crystal instead of particles and is made using a composite technique.[6,7] As the grain-aligned composite Terfenol-D consists of the preform, which maintains the bulk texture, and epoxy as an insulating layer, it is difficult to predict the magnetostriction of an epoxy-filled composite using a model based on magnetostrictive particles.

Herbst *et al.*[8] reported the effective magnetostriction of hot-pressed SmFe_2/Al (Fe) composites with SmFe_2 volume fractions of $f=0.2–0.8$ obtained from powdering annealed, single-phase ingots. They assumed that an elastically isotropic magnetostrictive sphere has a radius R_1 and saturation magnetostriction λ_s embedded in an elastically nonmagnetostrictive medium. Nan[9] studied the effective magnetostriction of the isotropic composite constituting a magnetostrictive particle and a nonmagnetostrictive phase using a Green function method. Chen *et al.*[10] investigated the effect of the volume fraction of various matrix materials (glass, glass+Al, Fe, Cu, or CeFe_2) on the magnetostriction of composites containing magnetostrictive particles. Guo *et al.*[11] studied the magnetostriction of glass, epoxy/Terfenol-D composites for V_f values of Terfenol-D particles in the range of 0.1–0.82 and suggested a model to describe the V_f dependence of magnetostriction and the coupling coefficient of the composite.

Since the reported magnetostrictive material model for a composite is based on the magnetostrictive particles, it is not necessary to consider the crystal orientation of the magnetostrictive phase. Therefore, relatively little research has examined the magnetostriction model of composite Terfenol-D, which maintains the texture of a unidirectionally solidified crystal. Here, we investigated the magnetostriction of grain-aligned composite Terfenol-D and developed a model to describe both the texture fraction and elastic modulus change with texture depending on the magnetostriction of the magnetostrictive phase ($R\text{Fe}_2$).

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II. EXPERIMENTAL PROCEDURES

$Tb_{0.33}Dy_{0.67}Fe_y$ alloys with different chemical compositions ($y = 1.10$, 1.36, and 1.66) were prepared using arc melting under high-purity argon and were then cast into rods using a suction method after induction melting. These rods were then sealed into a quartz ampoule and grown to a length of 150 mm by zone melting at a growth rate of 70 $\mu m/s$. For composite Terfenol-D, we first placed cylindrically machined samples (20 mm long and 6 mm in diameter) in a quartz ampoule containing quartz granules and sealed them at 360 Torr with purified argon to remove the eutectic phase from the as-grown Terfenol-D. On annealing at 1000 °C, the eutectic phase, which has a low melting point ($\sim$890 °C, although it differs slightly depending on the Tb/Dy ratio), leaked from the sample because of the reaction between the quartz granules and the sample. Then, the porous preform Terfenol-D, which is composed of cellular-structured RFe_2 prepared by extracting the eutectic phase, was immersed in liquid epoxy at a temperature of 70 °C, which has the lowest viscosity and pressurized with argon at 1200 Torr so that the porous preform with leaked channels was replaced with the epoxy. Finally, after curing the polymer infiltrated preform Terfenol-D, the grain-aligned composite Terfenol-D was finally fabricated using the unidirectional solidification technique.

The texture of the preform was measured by applying the Shultz reflection method using Cu $K\alpha$ and an orientation distribution function (ODF) analysis was performed to determine the texture information for the magnetostrictive phase (RFe_2) of the unidirectionally solidified crystal. Through ODF analysis, the volume fraction[12] of each direction with orientation g of the crystal axis with respect to a fixed sample coordinate system can be found from

$$F(g) = (V/dV)/dg. \tag{1}$$

Furthermore, the theoretical E_p (Ref. [13]) of preform Terfenol-D consisting of RFe_2 phase with a cubic structure is given by

$$1/E = S_{11} - 2(S_{11} - S_{12} - 1/2 S_{44})(l_1^2 l_2^2 + l_2^2 l_3^2 + l_3^2 l_1^2), \tag{2}$$

where s_{ij} and l_i are the compliance and directional cosine between each direction and the crystal axis (x, y, z), respectively. The fraction of RFe_2 phase in the composite was confirmed by measuring it with an image analyzer and determining the density. The magnetostrictive strain was measured along the longitudinal direction using a linear variable differential transform (LVDT) for each as-grown and grain-aligned composite Terfenol-D at room temperature. To measure the magnetostriction, a solenoid was used to provide a magnetic field of up to 80 kA/m and a compressive prestress ranging from 2 to 20 MPa was applied along the rod axis of the sample.[14]

III. RESULTS AND DISCUSSION

The magnetostriction model of grain-aligned composite Terfenol-D was based on the assumption of a uniform strain. Since in a preform of composite Terfenol-D prepared using unidirectional solidification the dendrite RFe_2 phase is ori-

ented in the longitudinal direction and because Poisson's ratios[15] of a preform Terfenol-D consisting of only the RFe_2 phase, and the epoxy in composite Terfenol-D are similar, it is reasonable to apply the uniform strain model. In the uniform strain model, the equilibrium of the forces in the longitudinal direction can be determined from

$$\sigma_c A_c = \sigma_{RFe_2} A_{RFe_2} + \sigma_e A_e, \tag{3}$$

where σ and A are the stress and cross-sectional area, respectively. The subscripts c, RFe_2, and e are the grain-aligned composite Terfenol-D, the magnetostrictive phase producing the magnetostriction, and epoxy with nonmagnetic properties, respectively. By inserting the strain of each component ($\varepsilon_c = \varepsilon_{RFe_2} = \varepsilon_e$) in Eq. (3), we get

$$E_c = E_{RFe_2} V_{RFe_2} + E_e V_e, \tag{4}$$

where E_c, E_{RFe_2}, and E_e are the elastic modulus of composite Terfenol-D, the RFe_2 phase, and epoxy, respectively.

In this magnetostriction model, because the magnetostriction in the grain-aligned composite Terfenol-D depends only on the magnetostrictive phase of RFe_2, we assume that the overall stress of the composite Terfenol-D as it is magnetically saturated, is the product of the fraction of the RFe_2 phase showing magnetostriction and the stress exerted by the RFe_2 phase. Under this condition, we can obtain

$$E_c \lambda_c = E_{RFe_2} V_{RFe_2} \lambda_{RFe_2}^{st}. \tag{5}$$

Since the grain-aligned composite Terfenol-D is based on preform Terfenol-D to maintain the texture of unidirectional solidification, Eq. (5) can be transformed into

$$E_c \lambda_c = \sum_{i=0}^{n} E_{h_i k_i l_i}^{RFe_2} V_{h_i k_i l_i}^{RFe_2} \lambda_{h_i k_i l_i}^{st}, \tag{6}$$

where E_{hkl} and V_{hkl} are the elastic modulus and volume fraction, respectively, for each direction of the preform Terfenol-D consisting of only the RFe_2 phase. Here, λ_{hkl} is the saturation magnetostriction for each direction in the RFe_2 phase. When we substitute the expressions for the elastic modulus in Eq. (4), we obtain the magnetostriction limiting value for the composite Terfenol-D as follows:

$$\lambda_c = \frac{\sum_{i=0}^{n} E_{h_i k_i l_i}^{RFe_2} V_{h_i k_i l_i}^{RFe_2} \lambda_{h_i k_i l_i}^{st}}{E_c}. \tag{7}$$

Since grains with different crystallographic orientations relative to the direction of measurement have different magnetostrictive properties, the various orientations in composite Terfenol-D consisting of a polycrystalline grown by zone melting should be considered to predict the effective magnetostriction of the grain-aligned composite Terfenol-D. Therefore, our magnetostriction model allows one to study the effect of the fraction of the magnetostrictive phase and the texture of the magnetostriction phase preserved during unidirectional solidification. The microstructure of grain-aligned composite Terfenol-D investigated in this model is given in Fig. 1, which shows that grain-aligned composite Terfenol-D is produced using the epoxy infiltration method.
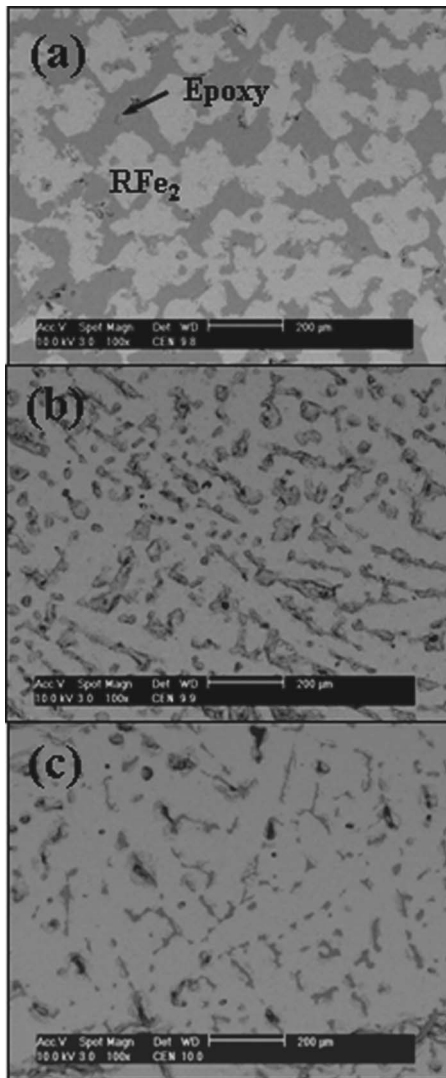
FIG. 1. Microstructures of grain-aligned composite Terfenol-D, following annealing and the infiltration of epoxy. Grain-aligned composite Terfenol-D with compositions of (a) $Tb_{0.33}Dy_{0.67}Fe_{1.10}$, (b) $Tb_{0.33}Dy_{0.67}Fe_{1.36}$, and (c) $Tb_{0.33}Dy_{0.67}Fe_{1.66}$.
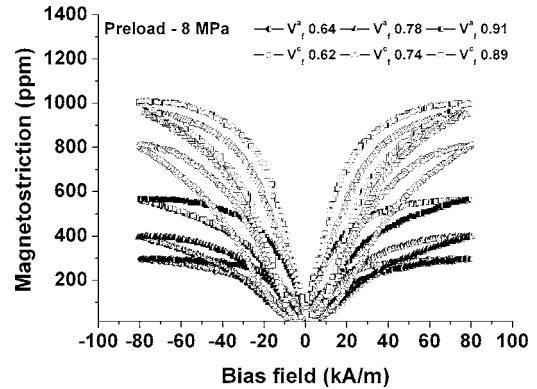

FIG. 2. Comparison of the magnetostriction of as-grown and grain-aligned composite Terfenol-D as a function of the bias field at 8 MPa. The superscripts c and a indicate composite and as-grown Terfenol-D, respectively.

The normalized fraction for each preferred direction of the magnetostrictive RFe_2 phase in grain-aligned composite Terfenol-D, the orientation distribution function $f(g)$ and the calculated elastic modulus of grain-aligned composite Terfenol-D are shown in Table I. The ODF results in Table I are very useful because they provide quantitative information about the entire spread of orientations in the texture. Based on this ODF, the normalized volume of the orientation space can be calculated by dividing the orientation density of each preferred orientation by the total ODF value.

In the elastic modulus calculation for grain-aligned composite Terfenol-D, the elastic modulus of the RFe_2 phase is calculated theoretically by summing the products of the calculated elastic modulus for each preferred direction and the normalized fraction of RFe_2 phase in each direction, while we use an elastic modulus value of 3.0 GPa for the epoxy. The theoretical elastic modulus of the RFe_2 phase is calculated using the elastic stiffness constants of monolithic Terfenol-D.[16] Moreover, when the RE/Fe ratio increases from 1.10 to 1.66, the preferred direction of the RFe_2 phase results in the transition from the $\langle 110 \rangle$ to the $\langle 111 \rangle$ direction.

The magnetostriction curves of the as-grown and grain-aligned composite Terfenol-D are presented in Fig. 2. This shows that the maximum magnetostriction of composite Terfenol-D is proportional to the fraction of the RFe_2 phase. When we remove the eutectic phase from as-grown Terfenol-D and then substitute epoxy for the eutectic phase, the magnetostriction of the grain-aligned composite Terfenol-D improves significantly (it is about two times higher than the magnetostriction of as-grown Terfenol-D). The improvement in the magnetostriction in the composite is divided into two broad contributions: annealing and the removal of the eutectic phase with nonmagnetic properties. The effect of the former on the magnetostriction was reported by Verhoeven *et al.*,[17] and Mei *et al.*,[18] who found that the magnetostriction improved to about 20% compared to the as-grown Terfenol-D during heat treatment due to relief of the residual stress in the as-solidified materials. Since the effect of heat treatment on magnetostriction in Terfenol-D is not major, the greatest improvement in the magnetostriction of the grain-aligned composite Terfenol-D compared to as-

TABLE I. Magnetostrictive phase fraction and normalized $f(g)$ value for as-grown Terfenol-D and the predicted elastic modulus of grain-aligned composite Terfenol-D.

Composition	Fraction of the RFe_2 phase		Normalized fraction of RFe_2/$f(g)$ value for each direction				Predicted elastic modulus of composite Terfenol-D (GPa)
	As grown	Composite	[110]	[113]	[112]	[111]	
$Tb_{0.33}Dy_{0.67}Fe_{1.10}$	0.64	0.62	0.52/5.6	0.22/3.5	0.26/3.5	⋯	26
$Tb_{0.33}Dy_{0.67}Fe_{1.36}$	0.78	0.74	0.45/6.3	0.25/4.1	0.30/4.0	⋯	37
$Tb_{0.33}Dy_{0.67}Fe_{1.66}$	0.91	0.89	0.56/14.8	⋯	0.16/5.1	0.28/7.8	56

TABLE II. The magnetostrictive properties of grain-aligned composite Terfenol-D. The maximum strain and d_{33} were obtained from measurements at 80 kA/m under a preload of 8 MPa, and the coupling coefficient was determined by measuring the impedance (direct method) considering the mass distribution.

Composition	Magnetostrictive properties (80 kA/m, 8 MPa)						Predicted magnetostriction of composite Terfenol-D (ppm)
	Max. strain (ppm)		d_{33} (nm/A)		k_{33}^{max}		
	As grown	Composite	As grown	Composite	As grown	Composite	
$Tb_{0.33}Dy_{0.67}Fe_{1.10}$	299.8	813.3	6.6	15.3	0.22	0.34	839.8
$Tb_{0.33}Dy_{0.67}Fe_{1.36}$	400.0	960.6	11.3	25.2	0.30	0.43	963.3
$Tb_{0.33}Dy_{0.67}Fe_{1.66}$	560.6	1013	13.4	27.8	0.36	0.45	1045.4

grown Terfenol-D at the same fraction of the $R Fe_2$ phase is largely attributable to the absence of the eutectic phase, which inhibits the magnetization rotations.

The measured maximum magnetostriction of as-grown and grain-aligned composite Terfenol-D and the predicted magnetostriction for the composite are shown in Table II. For the predicted magnetostriction of grain-aligned composite Terfenol-D, we used the saturation magnetostrictive strains of each orientation studied by Teter *et al.*[19] and Zhang *et al.*[20]

The magnetostriction model was developed as a function of the fraction of the magnetostrictive phase ($R Fe_2$) of grain-aligned composite Terfenol-D; this model used the elastic modulus obtained in the texture analysis for the magnetostrictive phase and 3 GPa for the epoxy. The results are compared to the experimental magnetostriction for the composite Terfenol-D in Fig. 3. In this figure, the open diamonds indicate the predicted magnetostriction based on the assumption of modified uniform strain. Furthermore, the dotted line shows the predictions for grain-aligned composite Terfenol-D under the uniform strain model, which does not consider the change in the elastic modulus with the change in the preferred direction of the crystal. Since several studies have reported that the preferred direction of monolithic Terfenol-D is $\langle 112 \rangle$,[18,19,21] in the uniform strain model, we assume that the crystal is completely oriented along the $\langle 112 \rangle$

direction, which has a saturation magnetostriction of 1640 ppm (Ref. 20) and that the elastic modulus of the magnetostrictive phase is 30 GPa (the mean value calculated from the wide range of stiffness values reported for Terfenol-D from 25 to 35 GPa). This uniform strain model indicates a major discrepancy compared to the experimental results for grain-aligned composite Terfenol-D. This suggests that the grain-aligned composite Terfenol-D produced using the crystal growth technique should consider the preferred direction of the magnetostrictive phase for grain-aligned composite Terfenol-D when determining the texture properties fabricated using zone melting method.

In this study, since our modified uniform strain model considered the change in the elastic modulus related to the crystal direction of the magnetostrictive phase, the predicted magnetostriction of the composite was in good agreement with the experimental results.

IV. CONCLUSION

In conclusion, we propose a model for predicting the effective magnetostriction of grain-aligned composite Terfenol-D, utilizing a modified uniform strain model that considers the elastic modulus coupled in the preferred direction. The predicted magnetostriction of grain-aligned composite Terfenol-D showed good agreement with the experimental results. Since our methodology accounts for the preferred direction of the unidirectionally grown crystal and the elastic modulus change related to each crystal direction, it can be used to assess the influences of different contributions on the effective magnetostriction. Therefore, this model can provide criteria for choosing the best combination of constituent materials for high-frequency magnetostrictive transducers or sensors. Furthermore, we confirmed that the effective magnetostriction of grain-aligned composite Terfenol-D depends on the preferred direction of the $R Fe_2$ phase.
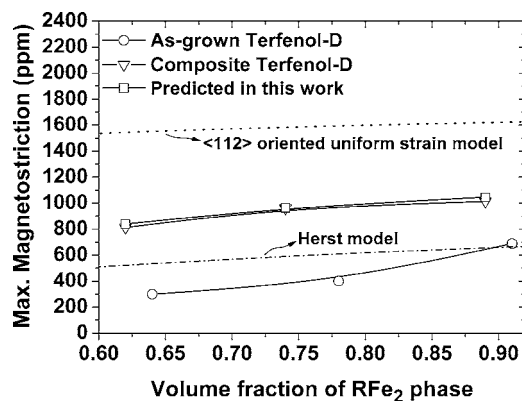

FIG. 3. Maximum magnetostriction and modeled results of the magnetostrictive strain calculated with the modified uniform strain condition considering the texture of the $R Fe_2$ phase and change in the elastic modulus with texture. The dotted line was calculated under the assumption that the magnetostrictive phase was oriented along the $\langle 112 \rangle$ direction. The dash-dotted line was calculated with the aid of the Herbst model (see Ref. 8), which assumes an elastically isotropic magnetostrictive sphere. The maximum magnetostriction was obtained at 80 kA/m and a preload of 8 MPa.

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