Magnetostriction and magnetomechanical coupling of grain-aligned 
Tb$_{0.33}$Dy$_{0.67}$Fe$_y$/epoxy-filled composites

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The magnetostriction and dynamic magnetomechanical coupling properties of the epoxy-filled composites were manufactured by replacing the eutectic phase with the epoxy were investigated as a function of the bias field and the RFe$_2$ volume fraction. The composites were prepared with RFe$_2$ volume fractions from 0.34 to 0.88 and were compared with as-grown crystals. The filled composites exhibit a greater maximum strain than do as-grown crystals, strongly dependent on the RFe$_2$ volume fraction. Furthermore, the dynamic magnetomechanical coupling coefficient of the filled composite, which was measured by resonance analysis, shows a maximum value of 0.45 at 47.2 kA/m, the highest known value for [1–3] composite Terfenol-D and strongly dependent on the volume fraction of RFe$_2$. © 2005 American Institute of Physics. [DOI: 10.1063/1.1925335]

I. INTRODUCTION

Giant magnetostrictive Terfenol-D can be employed as a transducer or actuator element up to approximately 10 kHz, but at higher frequencies, eddy current losses, which heat up the material and decrease the magnetic field penetrating the material, are known to restrict its applications. To minimize the restrictions imposed by monolithic Terfenol-D, such as eddy current loss and brittleness, some magnetostrictive Terfenol-D composites have been developed since the 1970s.1–3 In the initial stages, the magnetostrictive particulate [1–3] composites4,5 were made by integrating polycrystalline Terfenol-D particles dispersed in a nonmetallic binder such as epoxy, which created an insulating layer between the particles and thereby reduced eddy current losses at high frequencies. As this particulate [1–3] composite, however, is a combination of the demagnetization effect and the deficient strain transmission to neighboring particles, it produces only tiny strains of about 600–800 ppm at a magnetic field of 1 kOe. On the other hand, the particulate [1–3] composite reported so far, in which particles are aligned in one direction within the matrix, shows strains of about 1000 ppm at 12-MPa preload under 100 kA/m due to the connectivity of the particles and the significant reduction of the void.6,7

Recently, as the epoxy-filled Terfenol composite was introduced as an alternate composite, significantly improved magnetostriction and magnetostriective coefficients were obtained for frequency below about 10 kHz compared to the monolithic Terfenol-D.8–10 Although some studies have partially covered the dynamic magnetomechanical properties of particulate [1–3] composites, little research has been conducted on the dynamic magnetomechanical properties of the epoxy-filled Terfenol composite. Therefore, in this work, we investigated the magnetostriction and dynamic magnetomechanical properties of the epoxy-filled Terfenol composite.

and the dependence of the RFe$_2$ volume fraction on the magnetomechanical coefficient and the elastic modulus at a constant magnetic field.

II. MANUFACTURE PROCESS

The Tb$_{0.33}$Dy$_{0.67}$Fe$_y$ alloys, with the y values of 0.71, 0.90, 1.05, 1.36, and 1.65 composed of Tb, Dy, and Fe of high purity (99.99%), were prepared by arc melting under a high-purity argon and were then cast into rods via a suction method after induction melting. A directionally aligned cell structure of RFe$_2$ was obtained by zone melting at a growth rate of 70 µm/s, and the cast rods were then sealed into a quartz ampoule. To manufacture the composite, we put the cylindrically shaped samples into a quartz ampoule that contained quartz granules and sealed them at 360 Torr with a purified argon. The samples were then heated at a temperature of 1000 °C. During the reactive annealing at 1000 °C the eutectic phase, with a low melting point (~900 °C, though slightly different depending on the Tb/Dy ratio 11), reacted with the quartz granules (which are mainly composed of SiO$_2$) at the contact surface and leaked out of the sample. To permit sufficient wetting and to reduce voids in the composites, we immersed this porous preform, which is composed of cellular-structured RFe$_2$, into a liquid epoxy (as the binding material) at a temperature of 60 °C. After placing the preform in a vacuum for degassing, we pressurized it with argon at 1200 Torr. Then, after curing the preform, the composites were finally manufactured.

The volume fraction of RFe$_2$ in the composite was confirmed by measurement with an image analyzer. The magnetostrictive strain was measured along the longitudinal direction with linear variable differential transform (LVDT) at room temperature. A solenoid was used to provide a magnetic field of up to 80 kA/m, and a compressive prestress ranging from 2 to 10 MPa was applied along the rod axis of the sample. The magnetic induction was done simultaneously with the aid of a flux meter and a pickup coil, and the...
samples were demagnetized before each measurement. In addition, to determine the dynamic properties of the as-grown crystals and composites, we measured the impedance of the unloaded samples as a function of the bias field up to 80 kA/m. The bias field in the longitudinal direction of the samples was provided by the solenoid. We then wound the pickup coil tightly around the rods and placed the subassembly in a solenoid that generates a magnetic field from zero to 80 kA/m under an ac drive level of 40 A/m. The coupling coefficient \( k_{33} \) was determined by a direct method in which the mass distribution was considered.\(^{12}\) Its value is expressed as follows:

\[
k_{33}^2 = \frac{9}{8} \left[ 1 - \frac{f_R}{f_A} \right],
\]

where \( f_R \) and \( f_A \) are the resonance frequency and antiresonance frequency, respectively. In addition, the elastic modulus at a constant magnetic field was determined by using the resonance frequency \( f_R \) (Ref. 13), which is defined as

\[
f_R = \frac{1}{2L} \sqrt{\frac{E_{33}^H}{\rho}},
\]

where \( L \) is the length of the sample, \( \rho \) is the density of the sample (the average density as determined by the Archimedes principle), and \( E_{33}^H \) is the elastic modulus at a constant magnetic field. The antiresonance frequency is described by the same equation but with the constant magnetic induction \( E_{33}^B \), substituted for \( E_{33}^H \).

### III. RESULTS AND DISCUSSION

The microstructure in the \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \) alloy system is very sensitive to the RE/Fe ratio. As shown in Fig. 1(a), the as-grown microstructure reveals a dendrite RFe\(_2\) phase with the eutectic phase in between the dendrites. Figure 1(b) shows the preform the eutectic phase of which has been extracted from the as-grown crystal due to the reaction with SiO\(_2\) and the liquid phase. Figure 1(c) presents the epoxy-filled composite that was manufactured by substituting the eutectic phase with the epoxy. As shown in Fig. 1(c), the RFe\(_2\) volume fraction of the polymeric composites obtained by polymer infiltration method is a little bit lower than that of as-grown alloys shown in Fig. 1(c). This result indicates that at an annealing temperature of 1000 °C, not only is the eutectic phase extracted, but the primary RFe\(_2\) phase is also partly molten.

Figure 2 shows the dependence of the RFe\(_2\) volume fraction on the magnetostrictive strains as a function of the bias field. As the RFe\(_2\) volume fraction increases from 0.34 to 0.88, the magnetostrictive strain of the composite increases more significantly than the as-grown samples. The composite with a 0.88 volume fraction yields the highest magnetostrictive response of 1013 ppm. Since the magnetostriction in polycrystalline grown by float-zone melting was increased from 16% to 35% through heat treatment, as suggested by Verhoeven et al.,\(^{15}\) the increase in strain is too large to be due solely to the effect of annealing that reduces the pinning of the domain walls or enhances the rotation of domain moments through the residual stress relief of the as-grown crystal. That the magnetostriction in the composite is higher than that of the as-grown crystal at the same volume fraction of RFe\(_2\) is due to the absence of the eutectic phase. As suggested by Clark et al.,\(^{16}\) this result indicates that the eutectic phase inhibits the large magnetization rotations required for the giant magnetostrictive strain.

Figure 3 shows how the elastic modulus \( E_{33}^H \) depends on the bias field in the composites. The value of \( E_{33}^H \) reaches a minimum in all composites at a certain bias field value, and this increase is similar to the \( \Delta E \) effect in monolithic
Terfenol-D. The minimum $E^{III}_{33}$ appears at different bias fields which decrease as the volume fraction of the RFe$_2$ matrix in the composites increases from 0.34 to 0.88. This result means that since the $\Delta E$ effect results from magnetoelastic interaction (that is, there is an intrinsic softening of the crystal due to a local magnetoelastic atomic interaction$^{17}$), the magnetic moments rotate more easily at higher volume fractions of RFe$_2$.

Figure 4 shows the dependence of $E^{III}_{33}H$ on the volume fraction for all as-grown crystals and composites, as well as the analytical predictions of the elastic modulus. In this figure, the solid line represents the limiting theoretical values of the elastic modulus, which we calculated by assuming the isostrain condition$^{19}$ (the upper bound). For our calculation, we used an elastic modulus value of 3.0 GPa for the epoxy and a value of 51.4 GPa, which we measured by the resonance frequency method, for the annealed bulk Terfenol-D. The elastic modulus of the as-grown crystals decreases as the volume fraction of RFe$_2$ increases. In contrast, the elastic modulus of the composite increases as the volume fraction of RFe$_2$ decreases. For the as-grown crystals, as the volume fraction of the second phase decreases the enhancing fracture toughness, the elastic modulus of the crystal also decreases due to a reduction in the amount of the second phases with a higher stiffness. However, when we used an epoxy with a low elastic modulus to replace the eutectic phase, the elastic modulus in that composite increased as the amount of infiltrated epoxy decreased.

The experimental results are consistent with the isostrain condition, indicating an upper bound of the elastic modulus for the composites. Since the RFe$_2$ cells, which form a preform of a composite, are wellaligned along the longitudinal direction (the composites form 1-3 configuration when using the connectivity notation by Newnham$^{20}$ and the Poisson’s ratio of RFe$_2$ is 0.25, similar to that of the epoxy used in this experiment, we deduce that this model can be applied to the experimental results suggested by Chen et al.$^{21}$

The dependence of $k_{33}$ on the bias field for composites is shown in Fig. 5. The $k_{33}$ value initially increases to a maximum value; that is, $k_{33}$ is 0.45 for a composite with an RFe$_2$ volume fraction of 0.88 at 47.2 kA/m. This result means that the maximum fraction of the magnetic or elastic energies is transformed in the transduction process and decreases as the bias field increases. In addition, as shown in Fig. 3, the bias
field, where $k_{33}$ is maximized, is highly consistent with the value at which $E^H_{33}$ is minimized. This phenomenon is associated with the motion of domain walls from non-180° domain walls (that is, 70° domain walls\(^{23}\)) which reaches a maximum value, while the stiffness shows a minimum value. After $k_{33}$ reaches the maximum value, it gradually decreases due to the constraint of the non-180° domain wall motion. Furthermore, as the volume fraction of RFe\(_2\) in the composites increases from 0.34 to 0.88, the bias field, where the $k_{33}$ value is maximized, shifts to lower bias values. From these results, we deduce that composites with a lower volume fraction of RFe\(_2\) require additional energy to overcome the residual stress\(^{23}\) developed during the curing of the epoxy.

Figure 6 shows the experimental results of $k_{33}^{\text{max}}$ and the bias field where $k_{33}$ is maximized in relation to the volume fraction of RFe\(_2\) for the as-grown crystal and composites. The $k_{33}^{\text{max}}$ value in the as-grown crystals and composites increases as the volume fraction of RFe\(_2\) increases. Furthermore, since as-grown crystals are usually composed of two phases (the primary RFe\(_2\) phase and the eutectic phase, which consists of RFe\(_2\) and the rare-earth phase with a nonmagnetic phase\(^{24}\)), the coupling factor of the crystal falls with the increase of the eutectic volume fraction because the rare-earth phase within the eutectic is nonmagnetic. This phenomenon suggests that the nonmagnetic rare earth within the crystal causes local demagnetizing fields, which in turn cause the peak value of $k_{33}^{\text{max}}$ to shift to a higher field and decreases its magnitude. In the case of a composite, the value of $k_{33}^{\text{max}}$ is almost proportional to the volume fraction of RFe\(_2\), and the maximum bias field in which $k_{33}^{\text{max}}$ is maximized is inversely proportional to that of RFe\(_2\). In addition, at the same RFe\(_2\) volume fraction, the value of $k_{33}^{\text{max}}$ is double the corresponding value of the as-grown crystals. In this case, the higher coupling coefficient is due to the change in the elastic modulus of Eq. (1). Since the use of an epoxy with a lower modulus leads to an increase in $k_{33}^{\text{max}}$, we can observe that the energy-conversion efficiency of the composite is higher than that of the as-grown crystals in spite of the low drive field.

IV. CONCLUSION

In summary, the magnetostriction, elastic modulus, and magnetomechanical coupling of an epoxy and a Tb\(_{0.33}\)Dy\(_{0.67}\)Fe\(_2\) matrix composite combined with the unidirectional crystal growth technique and polymer infiltration method have been investigated as a function of volume fraction of RFe\(_2\). The results of this investigation have revealed that the epoxy strongly influences the magnetostriction and the magnetomechanical coupling properties. Furthermore, the magnetostriction and the magnetomechanical coupling coefficient greatly depend on the volume fraction of RFe\(_2\) within the preform. In this study when the volume fraction of the RFe\(_2\) was 0.88, the composite’s maximum magnetomechanical coupling $k_{33}^{\text{max}}$ was 4.5 at 47.2 kA/m. This coupling coefficient value is higher than those reported so far for composites made with [1–3] particles.

Furthermore, it is believed that the coupling coefficient of the composite depends on the elastic modulus of the filled material, such as epoxy, as well as on the volume fraction of RFe\(_2\).