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 that 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.
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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] composites[4,5] 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 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 magnetostrictive 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

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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:

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

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

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

where L is the length of the sample, ρ 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 $Tb_{0.33}Dy_{0.67}Fe_y$ 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.

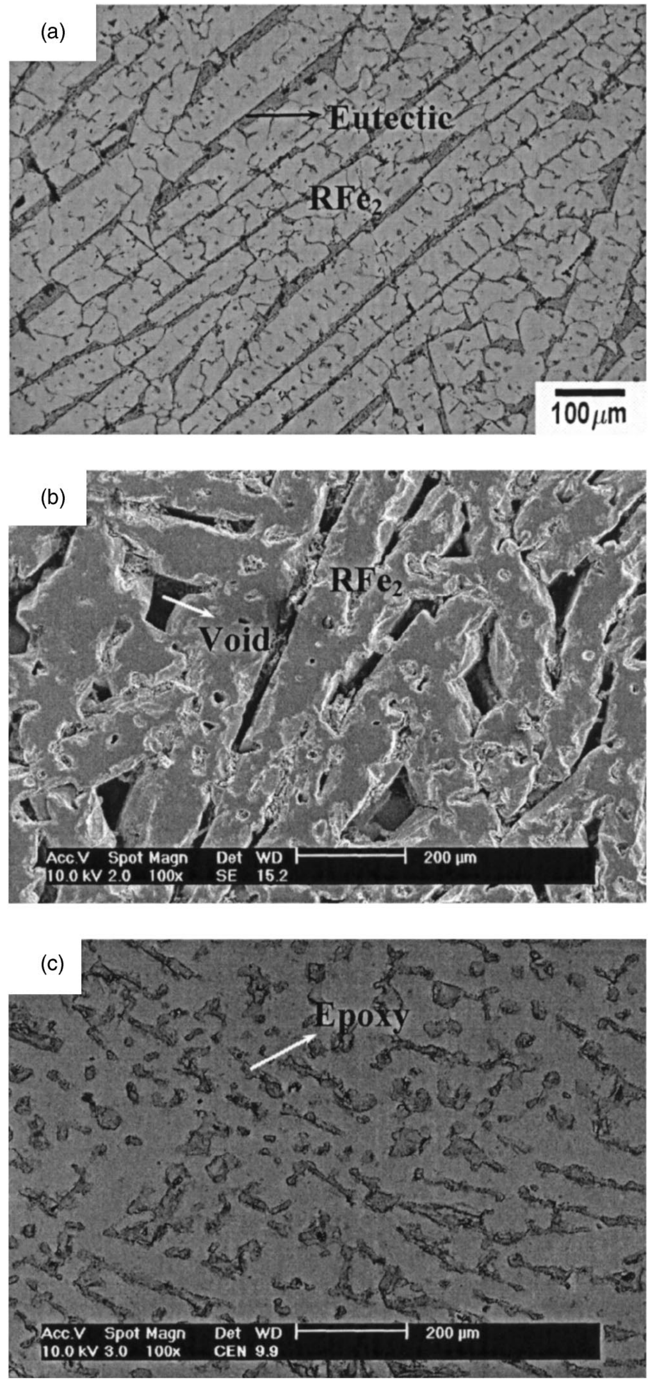

FIG. 1. Transverse sections for (a) the as-grown crystal, (b) the preform before making the composite, and (c) the filled composite with a composition of $Tb_{0.33}Dy_{0.67}Fe_{1.36}$ observed under optical or scanning electron microscope micrograph fitted with a backscattered electron.

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 mangetostriction 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 ΔE effect in monolithic

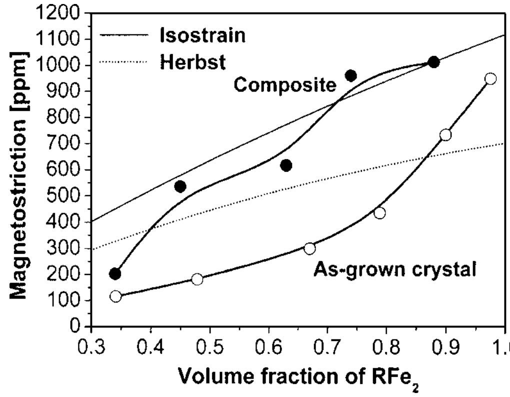

FIG. 2. Measured and modeled results of the magnetostrictive strain calculated with the isostrain condition assumption of filled composites with the epoxy phase of an elastic modulus and volume fractions of the RFe_2 phase (V_{RFe2}=0.34, 0.45, 0.63, 0.74, and 0.90). The dotted lines were calculated with the aid of the isostrain condition, and dash-dotted lines were calculated with the aid of the Herbst model (see Ref. 14), which assumes an elastically isotropic magnetostrictive sphere. The maximum magnetostrictive strains were obtained at 80 kA/m and a preload of 8 MPa.

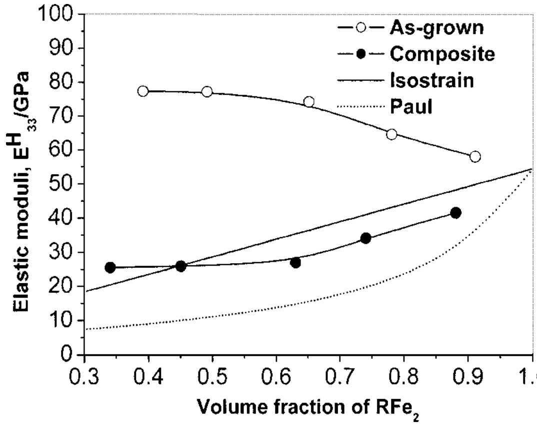

FIG. 4. Dependence of the minima of the E_{33}^H on the RFe_2 volume fraction in as-grown crystals and filled composites. The solid line was calculated by assuming an isostrain condition of E_{epoxy}=3 GPa, and E_{RFe2}=54.1 GPa; the dotted line is calculated by Paul's formula. (see Ref. 18).

Terfenol-D. The minimum E_{33}^H 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 ΔE 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_{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}^{max} 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

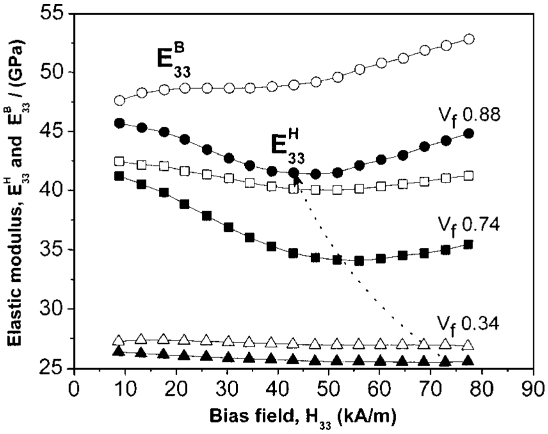

FIG. 3. Variation of E_{33}^H as a function of a bias field of filled composites with an RFe_2 volume fraction in the range of 0.34 to 0.88.

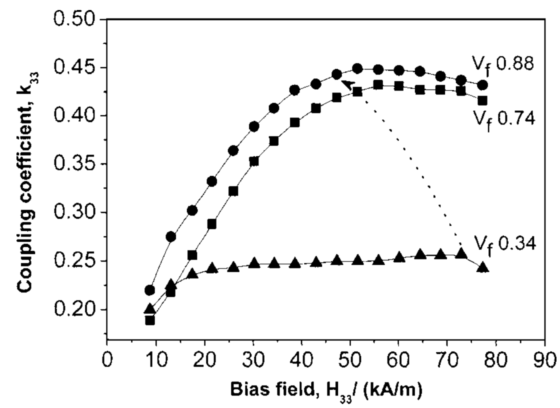

FIG. 5. The magnetomechanical coupling coefficient k_{33} as a function of the bias field for filled composites with RFe_2 volume fraction values ranging from 0.34 to 0.88.

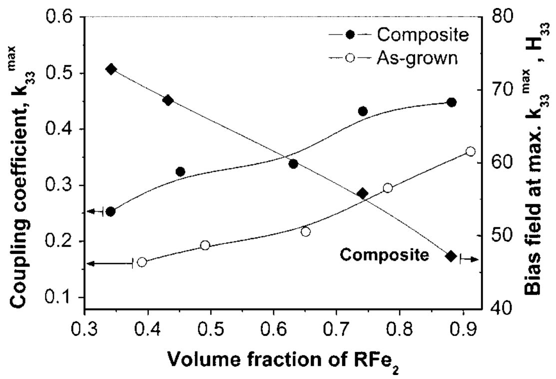

FIG. 6. The maximum coupling coefficient k_{33}^{max} and the bias field showing k_{33}^{max} as a function of the RFe_2 volume fraction for the as-grown crystals and filled composites.

field, where k_{33} is maximized, is highly consistent with the value at which E_{33}^{H} is minimized. This phenomenon is associated with the motion of domain walls from non-180° domain walls (that is, 70° domain walls[22]) 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 the 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}^{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}^{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}^{max} to shift to a higher field and decreases its magnitude. In the case of a composite, the value of k_{33}^{max} is almost proportional to the volume fraction of RFe_2, and the maximum bias field in which k_{33}^{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}^{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}^{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 $Tb0_{0.33}Dy_{0.67}Fe_y$ 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 magnetomehanical 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}^{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.

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