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Dynamic magnetomechanical properties of Terfenol-D/epoxy pseudo 1-3 composites

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Terfenol-D/epoxy pseudo 1-3 composites were fabricated by embedding and aligning Terfenol-D particles with a size distribution of 10–300 μm in a passive epoxy matrix using six Terfenol-D volume fractions (v_f) ranging from 0.22 to 0.72. The dependence of the dynamic relative permeability (μ_{r33}^T), elastic modulus (E_3^H), and dynamic strain coefficient (d_{33}) on v_f was investigated as a function of magnetic bias field (H_{Bias}). The H_{Bias} response data showed that the built-in non-180° domain states related to residual compressive stresses in the composites result in a significant decrease in μ_{r33}^T for $H_{\text{Bias}} < 40$ kA/m in addition to a minimization of E_3^H and a maximization of d_{33} near $H_{\text{Bias}} = 40$ kA/m. The v_f dependent data revealed that μ_{r33}^T is almost a linear function of v_f; E_3^H increases gradually with increasing v_f; and d_{33} increases initially, leveling off for $v_f > 0.5$. The present study provides a useful guide to optimize the composite properties for transducer design. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1851889]

Rare-earth-iron alloy Terfenol-D ($Tb_{0.30}Dy_{0.70}Fe_{1.92}$) is one of the finest magnetostrictive materials to date because of its giant magnetostrictive strain (~1200 ppm) and strain energy density (~20 kJ/m^3) with an expeditious response (~1 μs) at both room temperature and low fields (<150 kA/m). Nevertheless, two crucial problems have significantly limited its widespread use. The first is the limitation of the frequency to a few kilohertz due to the presence of eddy-current losses, while the second is difficulties in machining and fabricating devices owing to the brittleness of the material.

In recent years, it has been realized that magnetostrictive particulate composites based on Terfenol-D particles and a passive polymer binder can be fabricated to alleviate the problems intrinsic in monolithic Terfenol-D.[1] Additional benefits of using these composites are their tailorable properties and cost-effectiveness. Numerous studies have been conducted on two types of composites: namely, pseudo 1-3 composites (i.e., aligning Terfenol-D particles in a polymer matrix) and 0-3 composites (i.e., dispersing particles in the matrix).[1–6] In fact the early work undertaken by researchers aimed at maximizing the quasistatic (<10 Hz) strains through the optimization of composition parameters and the fabrication process. The dynamic behavior (i.e., frequency response data) of these materials remained unexplored until the early 2000s even though the critical concern has been to introduce the materials into high-frequency applications.

We have shown in pseudo 1-3 composites with a 0.5 Terfenol-D volume fraction ($v_f = 0.5$) that the eddy-current losses are insignificant for frequencies up to ~500 kHz, and the dynamic magnetomechanical properties, in particular E_3^H and d_{33}, are much higher than the 0-3 composites with the same v_f.[7] A further study discussed the influence of the combined magnetic bias (H_{Bias}) and drive (H_3) fields on the dynamic properties of this specific composite in terms of domain process.[8] To broaden the usage and to facilitate a proper use of this advanced material, improved understanding of the dynamic properties of the material on v_f is important. In this study, we aim to extend our work to investigate the dependence of both H_{Bias} and v_f on the dynamic properties of Terfenol-D/epoxy pseudo 1-3 composites. It is of great interest to obtain a minimal v_f that can generally provide an optimal set of property values for designing practical transducers.

Six batches of pseudo 1-3 composites with nominal v_f varying from 0.2 to 0.7 in steps of 0.1 were fabricated using irregular-shaped, 10–300 μm-sized, ball-milled Terfenol-D particles (Gansu Tianxing Rare Earth Functional Materials Co., Ltd., China) and Spurr epoxy (Polysciences, Inc., PA). For each batch of samples, predetermined quantities of Terfenol-D particles and epoxy were homogenously mixed in a bronze mold with a rectangular cavity of $12 \times 12 \times 30$ mm^3. The mixed slurry was degassed under a vacuum for 30 min to eliminate air bubbles. The mold was subsequently sealed to prevent particles from migration out once it was placed between a pair of NdFeB permanent magnets. These magnets produced a uniform magnetic field of ~150 kA/m along the longitudinal direction of the mold, causing the particles to align with the magnetic flux lines and producing chains similar to aligned short-fiber composites or, in general, a pseudo 1-3 composite. The entire mold-magnet assembly was placed in an oven at 70 °C for 8 h to ensure full cure of the epoxy and to introduce an average axial compressive stress of ~3 MPa in the composite. After demolding, the cured composite was cut and lapped into pieces with the desired dimensions of $5 \times 5 \times 25$ mm^3. Three samples were prepared for each v_f, and the average v_f of each batch of samples was determined to be 0.22, 0.32, 0.42, 0.51, 0.61, and 0.72 based on Archimedes' principle and rule-of-mixture formulation.

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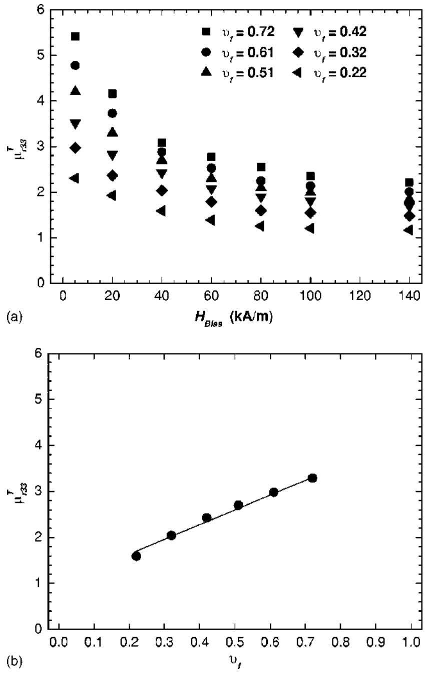

FIG. 1. (a) μ_{r33} at constant stress (μ^T_{r33}) measured at 1 kHz as a function of H_{Bias} for various υ_f and (b) υ_f dependence of μ^T_{r33} at $H_{\text{Bias}}=40$ kA/m, where the symbol and line represent the experimental and fitted data, respectively.

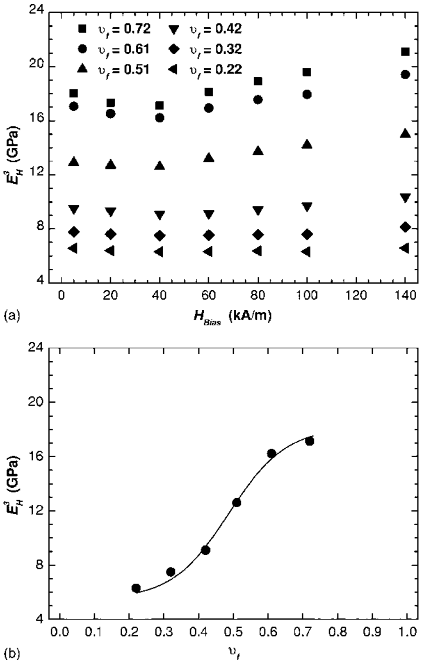

FIG. 2. (a) Dependence of E^H_3 on H_{Bias} for various υ_f and (b) variation of E^H_3 with υ_f at $H_{\text{Bias}}=40$ kA/m, where the symbol and line represent the experimental and fitted data, respectively.

The dynamic magnetomechanical properties of the samples in the longitudinal direction were measured at room temperature and with zero stress bias by sweeping a sinusoidal H_3 of 1 kA/m over a prescribed frequency range (f) of 1–100 kHz at a rate of 26 step/s and then measuring the corresponding magnetic flux density (B_3) and dynamic strain (S_3) at discrete frequency intervals of 25 Hz/step under various H_{Bias} of 5–240 kA/m. H_3 was provided by a Helmholtz coil driven by a dynamic signal analyzer (Ono Sokki CF5220) via a constant-current-supply amplifier (AE Techron 7572). H_{Bias} was supplied by a water-cooled, U-shaped electromagnet (Myltem PEM-8005K) controlled by a dc power supply (Sorensen DHP200-15). H_3 and H_{Bias} were monitored in-situ by a pick-up coil and a Gaussmeter (F. W. Bell 7030), respectively, while B_3 and S_3 were measured using a search coil wrapped around the sample and a strain gauge (Measurement Group EA-06-031CF-120-P) attached to the center of the sample and connected to a signal-conditioning amplifier (Measurement Group Vishay 2360), respectively. All quantities were sampled and recorded by the dynamic signal analyzer and stored in a computer. The dynamic relative permeability (μ_{r33}) was determined from

$$\mu_{r33} = \frac{B_3}{\mu_o H_3}, \tag{1}$$

where $\mu_o=4\pi\times10^{-7}$ H/m is the permeability of free space. The elastic modulus at constant magnetic field strength (E^H_3) was evaluated from the resonance (f_r) frequency as observed from the μ_{r33} spectrum by

$$E^H_3 = 4\rho(Lf_r)^2, \tag{2}$$

where L and ρ are the length and density of the composite, respectively. The dynamic strain coefficient (d_{33}) was obtained from

$$d_{33} = \frac{S_3}{H_3}. \tag{3}$$

Three samples were measured for each υ_f, and their average values are reported.

Figure 1(a) shows μ_{r33} at constant stress (μ^T_{r33}) measured at 1 kHz as a function of H_{Bias} for various υ_f. For all υ_f, μ^T_{r33} attains its maximum value at $H_{\text{Bias}}\leqslant 5$ kA/m and then decreases significantly in the H_{Bias} range of 5–40 kA/m before leveling off for $H_{\text{Bias}}>40$ kA/m. Since Terfenol-D particles have a cubic Laves phase, and their spontaneous magnetizations are essentially parallel to the $\langle 111\rangle$ easy axis, there exists a considerable amount of magnetic domains and domain walls in the particles. During the composite fabrication, residual axial compressive stresses are developed in composites while epoxy is cured. These built-in residual compressive stresses effectively create a preferred non-180° domain state in the composites as in the case of applying an external preload to assert an initial non-180° domain state in monolithic Terfenol-D.[2–5] Thus, the initial maximum in μ^T_{r33} at $H_{\text{Bias}}\leqslant 5$ kA/m is mainly attributed to the relatively easy 180° domain-wall motion. As H_{Bias} is increased beyond this level, the reduced 180° domain-wall motion competes with the increased non-180° domain-wall motion, resulting in a decrease in μ^T_{r33}. For $H_{\text{Bias}}>40$ kA/m, the contribution to μ^T_{r33} from the motion of 180° domain walls is negligible. This effect, together with constraining of non-180° domain-wall motion under H_{Bias}, tends to level off μ^T_{r33}. Accordingly, μ^T_{r33} exhibits a larger change from ~2.3 to ~5.4 at H_{Bias}

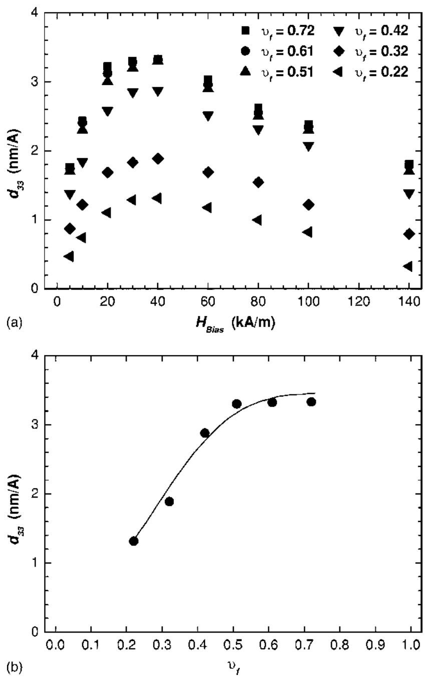

FIG. 3. (a) d_{33} measured at 1 kHz as a function of H_{Bias} for various υ_f and (b) υ_f dependence of d_{33} at $H_{\text{Bias}}=40$ kA/m, where the symbol and line represent the experimental and fitted data, respectively.

$=5$ kA/m in comparison with only a small change from ~1.2 to ~2.2 at $H_{\text{Bias}}=140$ kA/m when υ_f is increased from 0.22 to 0.72. The υ_f dependence of μ_{r33}^T at $H_{\text{Bias}}=40$ kA/m is plotted in Fig. 1(b). It is clear that μ_{r33}^T is almost a linear function of υ_f. Such a strict proportionality to υ_f serves to confirm the quality of the fabricated samples.

Figure 2(a) illustrates the dependence of E_3^H on H_{Bias} for various υ_f. All samples show an initial drop in E_3^H with increasing H_{Bias}. This is due to the H_{Bias}-induced motion of the available non-180° domain walls. As H_{Bias} is increased near 40 kA/m, the compliance associated with increased deformation contribution from this non-180° domain wall motion is maximized, leading to a minimum in E_3^H. Beyond this H_{Bias} level, E_3^H as a function of H_{Bias} displays an increasing trend. The effect is characterized by constraining of non-180° domain-wall motion due to interaction with H_{Bias}. Figure 2(b) shows the variation of E_3^H with υ_f at $H_{\text{Bias}}=40$ kA/m. While there is a slow increase in E_3^H for both the low (<0.4) and high (>0.6) υ_f regions as compared with a more evident increase in the medium υ_f region (0.4–0.6), E_3^H exhibits a gradual increasing trend with υ_f in general. The fact that E_3^H experiences a more rapid increase in $\upsilon_f=0.4-0.6$ may be explained by the effect of particle alignment in the composites against different υ_f. At a sufficiently low υ_f value (~0.4), the embedded number of particles is very limited, and the particles are not physically in touch with each other even though they are intentionally aligned as continuous chains in a macroscopic view. At an adequately high υ_f value (~0.6), there are a sufficient number of particles for collocating continuous chains with a sufficiently high packing density. Thus, there exists a transition between these two extremes (i.e., $\upsilon_f=0.4-0.6$).

Figure 3(a) shows d_{33} at 1 kHz as a function of H_{Bias} for various υ_f. d_{33} of all samples increases initially and displays a maximum near $H_{\text{Bias}}=40$ kA/m. This is a result of increasing and maximizing S_3 contribution from the non-180° domain-wall motion, respectively. In particular, the occurrence of maximum d_{33} at ~40 kA/m suggests that the composites are biased in the center of the "burst region" of their quasistatic strain-field curves.[7–9] The results agree with the initial decrease in both μ_{r33}^T and E_3^H for $H_{\text{Bias}}<40$ kA/m [Figs. 1(a) and 2(a)], since the initial 180° domain-wall motion produces changes in magnetization (and hence μ_{r33}^T) without accompanying S_3 while the later non-180° domain-wall motion produces changes in S_3 rather than changes in μ_{r33}^T.[10] Above this critical H_{Bias} level, the decrease in d_{33} with increasing H_{Bias} results from domain saturation, where S_3 remains essentially constant with increasing H_{Bias}. The behavior of d_{33} at $H_{\text{Bias}}=40$ kA/m is plotted against υ_f in Fig. 3(b). d_{33} increases monotonically up to $\upsilon_f=0.5$ and then remains almost constant with further increasing υ_f. This indicates that the use of composites with $\upsilon_f=0.5$ is sufficient for producing an optimal d_{33} value. This monotonic increase in d_{33} with increasing υ_f also reveals that more of the stress on the faces of the composites is borne by the Terfenol-D chains and less by the epoxy matrix as υ_f increases. Since Terfenol-D is much stiffer than epoxy, d_{33} of composites attains almost the value of the polycrystalline Terfenol-D's d_{33} already at low υ_f (~0.5).

It has been shown that μ_{r33}^T, E_3^H, and d_{33} of Terfenol-D/epoxy pseudo 1-3 composites are essentially a function of both H_{Bias} and υ_f. μ_{r33}^T reaches its minimum value at $H_{\text{Bias}} \leqslant 5$ kA/m due to the relatively easy 180° domain-wall motion, while both E_3^H and d_{33} exhibit a minimum and a maximum around $H_{\text{Bias}}=40$ kA/m, respectively as a result of the maximum motion of the built-in non-180° domain-walls. For use in practical transducers, the composites should have a reasonably high μ_{r33}^T, a moderately high E_3^H, a high d_{33}, and a sufficiently low cost. Therefore, the optimal device performance and cost can be obtained by using composites of υ_f ~0.6.

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