Stress-Rate Dependent Output Voltage for Fe₂₉Co₇₁ Magnetostrictive Fiber/Polymer Composites: Fabrication, Experimental Observation and Theoretical Prediction

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The Villari effect of magnetostrictive materials, a change in magnetization due to an applied stress, is used for sensor/energy harvesting applications. In this work, magnetostrictive fiber/polymer composites are fabricated for the first time by embedding strong textured Fe–Co fibers in an epoxy matrix, and their stress-rate dependent output voltage characteristics are investigated. Compression tests are first conducted to measure the output voltage of a sample. A simple magnetomechanical coupling model of the magnetostrictive fiber/polymer composite is then established. The output voltage is predicted, and domain wall dynamics is discussed in relation to the macroscopic inverse magnetostrictive response (known as the Villari effect). The results show that the output voltage density of this novel Fe–Co fiber/polymer composite dramatically increases with increasing stress-rate and becomes larger than that of Fe–Ga alloy. Our work represents an important step forward in the development of magnetostrictive sensor and energy harvesting materials. [doi:10.2320/matertrans.M2016410]

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1. Introduction

Energy harvesting technologies are recently receiving significant interest for wireless sensor and internet of things (IoT) applications. The use of inverse magnetostriction is a possible approach for energy harvesting, and low cost materials with high magnetostriction are required. Among magnetostrictive materials, Terbium Dysprosium Iron alloy (Terfenol-D) is the most attractive one due to its giant magnetostriction (800–1600 ppm) and low magnetic anisotropy¹⁾. Recently, Mori *et al.*²⁾ studied the output voltage characteristics of Terfenol-D cantilevers with resonant tuning both numerically and experimentally. Although Terfenol-D is potentially important as an energy harvesting material, several issues such as brittleness and the occurrence of high eddy currents have limited the adoption of this material. Polymer matrix composites containing Terfenol-D particles produce a relatively tough material, and the polymer creates an insulating layer between magnetostrictive particles and eliminates eddy current losses³⁾. Yoffe *et al.*⁴⁾ introduced a new procedure for modeling the magnetic field induced by an external load applied to epoxy-based composites with Terfenol-D particles. Although these magnetostrictive composites have attracted considerable interest, Terfenol-D particle is a rare earth iron alloy and very expensive.

Fe–Ga alloys (Galfenol) exhibit magnetostriction of 400 ppm, and some researchers investigated the energy harvesting characteristics of Galfenol vibration power generators^{5,6)}. However, they display some drawbacks such as difficulty in production. On the other hand, Fe–Co alloys can be good candidate magnetostrictive materials applicable to the energy harvesting devices due to their abundance and lower cost compared with Terfenol-D and Galfenol. Recently, magnetostriction of Fe_{1–x}Co_x (x = 50–90 at%) alloys prepared by forging and subsequent cold rolling was studied for various alloy compositions and thermomechanical treatments by Ya-

maura *et al.*⁷⁾ Fe–Co alloys exhibit high strength, ductility, and excellent workability, allowing easy fabrication of Fe–Co wires, and Narita⁸⁾ developed the magnetostrictive wire/polymer composites that exploit the characteristics of Fe₂₉Co₇₁ wires with diameter of approximately 1 mm, and elucidated the stress-rate dependence of the output voltage of the composites due to compression.

It is expected that the embedding of thinner Fe–Co fibers in a polymer will lead to magnetostrictive composite materials having more enhanced magnetostriction. In this study, magnetostrictive fiber/polymer composites are developed by embedding Fe₂₉Co₇₁ fibers with diameter of approximately 200 μm in the epoxy matrix, and the stress-rate dependent output voltage characteristics are discussed theoretically and experimentally.

2. Experimental Procedure

Fe_{1–x}Co_x (x = 71 at%) wire with a diameter of 1 mm was prepared as a filler. Wire drawing was executed to obtain the strong textured high strength fiber of approximately 200 μm diameter (see Fig. 1(a)). Composite with no prestress was then fabricated with the Fe₂₉Co₇₁ fibers combined with a commercially available epoxy resin based on diglycidyl ether of bisphenol-F with a polyamine curing agent (see Fig. 1(b)). 125 fibers with a length of approximately *l* = 19 mm were cut.

Fig. 1 Optical images of (a) Fe₂₉Co₇₁ fiber by drawing, (b) composite sample and (c) cross-section.

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The easy axis is in the fiber direction. These fibers were unidirectionally aligned inside a mold. The epoxy resin was transferred into the mold, cured for 1 day at room temperature, and cured for 3 h at 80°C inside an oven. The cross-sectional area of the composite sample is $A = 69.5 \text{ mm}^2$ (see Fig. 1(c)). The fiber volume fraction is $v^f = 0.0565$, where the superscript f indicates the fiber.

Uniaxial cyclic compression tests were performed on the composite sample loaded in Autograph (SHIMAZU AG-50kNXD). At each testing step, the sample was cyclically loaded and unloaded five times to the same magnitude of the maximum stress $\sigma_{\max} = P_{\max}/A$ (where $P_{\max}$ is the maximum load) while permanent magnets were attached directly on the bottom of the sample to apply a magnetic bias field $B_0 = 118 \text{ mT}$. The forces were driven with constant crosshead velocities of $d\delta/dt = 0.25, 0.50, 0.75, 1.0, 2.0$ and 3.0 mm/s (where δ is the crosshead displacement and t is the time). The output voltage V_{out} of the sample was measured with a search coil and a data logger. The coil had a circular cross-sectional area of approximately 113 mm^2 and a length of approximately 36 mm with 2500 turns.

3. Theoretical Analysis

Here, we model the representative volume element (RVE) as a single cylindrical Fe–Co fiber bonded within a cylinder of the epoxy matrix. The ratio of the Fe–Co fiber radius and epoxy cylinder radius is $(v^f)^{1/2}$; the fiber volume fraction within the RVE is v^f , which is the same as the fiber volume fraction for the entire sample.

We now consider the system using rectangular Cartesian coordinates x_i ($O-x_1, x_2, x_3$). The easy axis for the magnetization of the Fe–Co fiber is along the x_3 -direction. The basic equations can be found in Ref. 2). When the long dimension of the composite is much longer than the diameter and the magnetic bias field is along the long direction (x_3 -axis), the longitudinal (33) magnetostrictive deformation mode is dominant. Therefore, it is assumed that the magnetoelastic constant varies with the x_3 -component of the magnetic field intensity vector⁹⁾. In this case, it can be treated as a one-dimensional problem, and therefore, the constitutive laws of the Fe–Co fiber can be written as

$$\begin{aligned}\varepsilon_{33}^f &= s_{33}^f \sigma_{33}^f + (d_{33}^f + m_{33}^f H_3^f) H_3^f, \\ B_3^f &= (d_{33}^f + m_{33}^f H_3^f) \sigma_{33}^f + \mu_{33}^f H_3^f,\end{aligned}\quad (1)$$

where σ_{33}^f and ε_{33}^f are the components of the stress and strain tensors, and B_3^f are H_3^f the components of the magnetic induction and magnetic field intensity vectors, s_{33}^f is the elastic compliance at a constant magnetic field, d_{33}^f is the piezomagnetic constant, m_{33}^f is the second-order magnetoelastic constant, and μ_{33}^f is the magnetic permeability at constant stress, respectively. The material properties of the Fe₂₉Co₇₁ fiber are $s_{33}^f = 5.5 \times 10^{-12} \text{ m}^2/\text{N}$, $d_{33}^f = 0.125 \times 10^{-9} \text{ m/A}$, $m_{33}^f = 0.0123 \times 10^{-12} \text{ m}^2/\text{A}^2$ and $\mu_{33}^f = 37.7 \times 10^{-6} \text{ H/m}$. Assuming that the induced magnetic field intensity due to the inverse magnetostrictive effect is very small compared with the magnetic bias field $H_0 = B_0/\mu_0$ (where $\mu_0 = 1.26 \times 10^{-6} \text{ H/m}$ is the magnetic permeability of free space), eq. (1) can be simply expressed as

$$\begin{aligned}\varepsilon_{33}^f &= s_{33}^f \sigma_{33}^f + d_{33}^f \left(\frac{B_0}{\mu_{33}^f} \right) + m_{33}^f \left(\frac{B_0}{\mu_{33}^f} \right)^2, \\ B_3^f &= d_{33}^f \sigma_{33}^f + m_{33}^f \left(\frac{B_0}{\mu_{33}^f} \right) \sigma_{33}^f + B_0.\end{aligned}\quad (2)$$

The strain tensor component ε_{33}^m and magnetic induction vector component B_3^m for the epoxy matrix are given by

$$\varepsilon_{33}^m = s_{33}^m \sigma_{33}^m, \quad B_3^m = \mu_0 H_3^m, \quad (3)$$

respectively, where σ_{33}^m is the component of the stress tensor, H_3^m is the component of the magnetic field intensity vector, and s_{33}^m is the elastic compliance of the matrix. The superscript m denotes the matrix. The material property of the epoxy is $s_{33}^m = 400 \times 10^{-12} \text{ m}^2/\text{N}$.

The perfect bonding between the Fe–Co fiber and epoxy cylinder impose that $\varepsilon_{33}^f = \varepsilon_{33}^m$. Due to the transverse isotropy of the RVE, σ_{33}^f and σ_{33}^m are uniform and therefore the average stress can be given by

$$\sigma_{33}^0 = \sigma_{33}^f v^f + \sigma_{33}^m (1 - v^f). \quad (4)$$

This stress σ_{33}^0 is equivalent to the stress imposed by the loading machine on the overall sample. The output voltage of the composite is also obtained as

$$\begin{aligned}V_{\text{out}} &= -NA^f \frac{dB_3^f}{dt} \\ &= -NA^f \frac{s_{33}^m}{s_{33}^m v^f + s_{33}^f (1 - v^f)} \left\{ d_{33}^f + \left(\frac{m_{33}^f}{\mu_{33}^f} \right) B_0 \right\} \frac{d\sigma_{33}^0}{dt},\end{aligned}\quad (5)$$

where N is the number of turns in the search coil and A^f is the cross-sectional area of the Fe–Co fiber.

On the other hand, a large domain wall velocity v_w induces the following local magnetic induction change¹⁰⁾:

$$\frac{dB_3^f}{dt} \propto \alpha \mu_0 M_s \frac{v_w}{l_w}, \quad (6)$$

where M_s is the saturation magnetization, l_w is the domain wall jumping length related to the grain size, and α is a constant related to the susceptibility and number of jumping walls.

4. Results and Discussion

Figure 2 shows the measured output voltage density V_{out}/Alv^f (average values of five data), with error bars indicating standard deviation, versus stress-rate $d\sigma_{33}^0/dt$ and the maximum stress $\sigma_{\max}$ for the loaded and unloaded states of the Fe₂₉Co₇₁ fiber/polymer composite. For comparison, the previous data⁸⁾ for the composite with Fe₂₉Co₇₁ wires of 1 mm diameter and Galfenol bulk are shown. Note that high output voltage density is observed for the loaded state of the Fe₂₉Co₇₁ fiber/polymer composite for the following three reasons. First, this is because of the fact that the applied compressive stress in the fiber becomes very high. For example, under the average stress $\sigma_{33}^0 = \sigma_{\max} = 50 \text{ MPa}$, the corresponding compressive stress σ_{33}^f in the Fe₂₉Co₇₁ fiber of the RVE ($v^f = 0.0565$) can be calculated to be approximately 720 MPa, and it is presumed that this stress enhanced the magnetic flux leakage (see the second of eq. (2)). Second, using variations

Fig. 2 Measured output voltage density versus stress-rate and maximum stress for $\text{Fe}_{29}\text{Co}_{71}$ fiber/polymer composite, $\text{Fe}_{29}\text{Co}_{71}$ wire/polymer composite⁸⁾ and Galfenol bulk⁸⁾.

Fig. 3 SEM images of the surfaces for (a), (b) $\text{Fe}_{29}\text{Co}_{71}$ fiber, (c) $\text{Fe}_{29}\text{Co}_{71}$ wire⁸⁾ and (d) Galfenol bulk.

in processing, we demonstrate a material with high crystallinity and magnetic anisotropy. Third, because of the high aspect ratio of the fibers, the effect of the demagnetization fields is effectively reduced. Figure 3 shows the scanning electron microscope (SEM) images of the surfaces of the $\text{Fe}_{29}\text{Co}_{71}$ fiber, $\text{Fe}_{29}\text{Co}_{71}$ wire⁸⁾ and Galfenol bulk. A strong texture can be found in the fiber. This means that the superior texture was obtained in the fiber direction after the wire drawing.

Figure 4 shows the measured and calculated output voltage density V_{out}/Alv^f versus stress-rate $d\sigma_{33}^0/dt$ for the unloaded state of the $\text{Fe}_{29}\text{Co}_{71}$ fiber/polymer composite under the maximum stress $\sigma_{\text{max}} \approx 8$ MPa. The trend is sufficiently similar between the calculation (eq. (5)) and measurement. However, differences exist between theoretical and experimental results. This distinction can be attributed to the Barkhausen

Fig. 4 Measured and calculated output voltage density versus stress-rate for $\text{Fe}_{29}\text{Co}_{71}$ fiber/polymer composite.

voltage related to eq. (6). From the perspective of domain wall dynamics, we found out that microstructural factor such as domain wall jumping length " l_w " contributes to the macroscopic inverse magnetostrictive response.

5. Conclusion

Novel magnetostrictive composite materials, utilizing the characteristics of the strong textured $\text{Fe}_{29}\text{Co}_{71}$ fiber, were developed for the first time. The output voltage increased with increasing stress-rate. As a result, the new composite material exhibits properties that surpass the properties of Galfenol. Here we used a non-prestressed composite, but if we can introduce prestress into the fiber, it will likely produce much greater output voltage. This work opens the door for development of lightweight, robust, and efficient sensor and energy harvesting materials.

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