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Enhanced magnetoelectric efficiency of the $Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3$ cylinder multi-electrode composites

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ABSTRACT

Cylinder layered magnetoelectric (ME) $Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3$ (T–P), $Pb(Zr,Ti)O_3/Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3$ (P–T–P) and $Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3/Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3$ (T–P–T–P) composites with the same overall dimensions, but different structures and layers were designed and prepared for enhancing magnetoelectric efficiency. The ME effect in the vertical mode was studied. Compared with the T–P and T–P–T–P composites, the hollow P–T–P composite with a multi-electrode cylinder structure has higher ME effect and uses less $Tb_{1-x}Dy_xFe_{2-y}$ (Terfenol-D, a piezomagnetic material). The ME efficiency factor of P–T–P is more than two times higher than the other two composites. The reason for the improvement can be ascribed to the effective working surface conditions and the series multi-electrode effect. The results of the study indicate that this type of hollow multi-electrode cylinder ME composite can enhance the efficiency of piezomagnetic materials, particularly Terfenol-D. This finding demonstrates that creating similar structural designs not only reduces the weight, but also provides a cost effective way to manufacture ME devices for practical applications.

1. Introduction

Magnetoelectric (ME) composites made by combining piezoelectric (PE) and piezomagnetic (PM) materials can lead to excellent ME effect at room temperature, compared with the single phase magnetoelectrics [1]. The ME effect is defined as the induced dielectric polarization under applied magnetic field (H) through interfacial strain coupling of the two phases [1,2]. ME composite materials, particularly layered composites made by combining PM and PM layers, have been intensively studied for their promising applications in multifunctional devices due to a simple manufacturing process [3], including, but not limited to transducers, actuators and sensors [4–7]. Due to potential applications [8,9], ME composites have become a popular research topic, being an integral part of the devices design [10–12].

Layered ME composites that display remarkable performance contain high quality piezoelectric (PE) materials [13,14], such as $Pb(Zr,Ti)O_3$ (PZT) and $Pb(Mg_{1/3}Nb_{2/3})_{(1-x)}Ti_xO_3$ (PMN–PT) and piezomagnetic (PM) materials, such as $Tb_{1-x}Dy_xFe_{2-y}$ (Terfenol-D) [15]. Currently, Terfenol-D is one of the best PM phase candidates for the ME composites with giant magnetostrictive capabilities [16,17]. Zuo [18] prepared Terfenol-D/PZT multi-electrode laminated composites produced by bonding square shaped PZT and a thick layer of Terfenol-D with epoxy, resulting in an excellent voltage coefficient (α_E) of about 24 V/cm·Oe. However, the composite needs a relatively thick layer, weighing 29 g, of Terfenol-D under the DC magnetic bias (H_{DC}) to induce dielectric polarization of PZT. Pan [19] reported that monocyclic cylinder structural ME composites have better ME effect than monocyclic laminated ones with the same effective dimensions. Most of the current studies of the ME composites mainly focus on higher ME voltage coefficients, but usually do not focus on the amount or weight of the materials used. PM materials, such as Terfenol-D are fairly expensive due to the rare-earth alloys, terbium (Tb) and dysprosium (Dy) components, which make up more than half of the Terfenol-D alloy [20]. Creating an optimized structural design to utilize PM materials in an efficient way, namely by enhancing the magnetoelectric efficiency factors, would allow PM materials to contribute more power (ME effect) per unit weight [21]. Therefore, it would be more beneficial to create more efficient lightweight devices for reducing the cost of devices and applications. In order to attain both high ME effect and less PM material usage, multi-electrode cylinder layered ME composites PZT/Terfenol-D/PZT (P–T–P), based on the advantages of series mode laminated Terfenol-D/PZT composite and cylinder layered Ni/PZT composite, were prepared by compression molding with epoxy [6,18]. Furthermore, a basic structural monocyclic Terfenol-D/PZT (T–P) composite with a single electrode layer and a Terfenol-D/PZT/Terfenol-D/PZT (T–P–T–P) composite with the same overall dimensions (same two layers

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of electrode PZT structures, but more Terfenol-D usage than the P–T–P), were prepared for comparison. The vertical mode voltage coefficients ($\alpha_{E,V}$) of the samples were investigated.

2. Materials and methods

The proposed layered ME composites, as shown in Fig. 1, were made up of a positive magnetostrictive phase Terfenol-D and piezoelectric PZT phase. The PZT cylinders were polarized along the radial direction after electroplating a thin layer of Ni on the inside and outside surfaces. The Terfenol-D particles with ~180 μm size were obtained by crushing bulk Terfenol-D single crystal in argon. A homogeneous mixture consisting of 12 wt.% epoxy resin binder and 88 wt.% Terfenol-D particles was compacted by uniaxial pressing with randomly oriented particles. Terfenol-D-epoxy needed 24 h for curing. The ME effect of the Terfenol-D/PZT composites was obtained using the ME measurement system, where constant (H_{DC}) and alternating (δH) magnetic fields were applied in the vertical mode. The ME voltage coefficient was calculated as $a_E = \delta V/(t_{PE} \cdot \delta H)$, where t_{PE} is the PZT layer thickness and δH is the amplitude of the AC magnetic field generated by the Helmholtz coils. The AC current flowing through the coil with the applied magnetic field amplitude of $\delta H = 1.2$ Oe was equal to 1 A, where δV is differential voltage from the differential applied magnetic field (δH). The output voltage δV across the sample was measured via an oscilloscope. The vertical mode ME voltage coefficient, $\alpha_{E,V}$, was measured when H_{DC} and δH were applied along the vertical direction of the cylinder, as shown schematically in Fig. 2(a). For comparison, the T–P, P–T–P and T–P–T–P cylindrical layered composites were prepared. Both P–T–P and T–P–T–P have two layers of PZT connected in series [18] (inner and outer PZT electrodes connected in series). The geometrical arrangements and components of three composite samples are shown schematically in Fig. 1.

3. Results and discussion

Fig. 2(a) illustrates the directions of the applied magnetic field and polarization of the composite in the vertical mode (P–T–P is shown as an example). Fig. 2(b) shows the ME voltage coefficient, $\alpha_{E,V}$, dependency on frequency in the vertical mode for the three Terfenol-D/PZT cylinder layered composites at the respective optimal magnetic field (H_m). As shown in Fig. 2(b), each composite has one main resonance peak of $a_{E,V}$, each having relatively close resonant frequencies. The slight difference of the three resonant frequencies is related to the electromechanical resonance of the three samples with relatively different structures and mass [22]. Fig. 2(c) shows the H bias (H_{DC}) dependence of the ME coefficient ($a_{E,V}$) at the respective resonant frequency. The $\alpha_{E,V}$ increased with H_{DC} until the local maximum value was reached for the three tested composites. By comparison, the P–T–P composite showed that the maximum $a_{E,V}$ value of 5.8 V/cm·Oe is higher than that of the other two composites, T–P and T–P–T–P.

Fig. 3 shows a schematic illustration of the contact surfaces (cyan area) and the free surfaces (dark gray area) of the three composites without PZT. The contact surface is defined as the contact area of Terfenol-D and PZT, while the other areas are the free surfaces [19]. As shown in Fig. 3, each composite has different contact and free areas. The total area of contact surfaces and free surfaces of each composite was calculated using the circle area ($A_{circle} = \pi \cdot r^2$) and the cylinder area ($A_{cylinder} = \pi \cdot D \times t$), where A is the area, r is the radius, D is the diameter, and t is the thickness of the composite. An effective working surface specific value (A_{eff}) is calculated by dividing the area of contact surface by the area of the free surface for each composite. In comparison, the P–T–P has the largest A_{eff} value of 1.25, whereas T–P A_{eff} is equal to 0.43 and T–P–T–P A_{eff} is equal to 1.08.

According to the Pan's differential coefficient model [23], a vertical coupling mode cylindrical composite can be simplified as an effective 2–2 laminated ME composite. In the vertical mode, both $\alpha_{E,31}$ and $\alpha_{E,33}$ of this effective ME composite contribute to the ME effect ($a_{E,V}$) at the same time [23]. Based on the constitutive equation [16], when a perfect interface between the PM and PE phases exists in an ideal case, and a magnetic field is applied along the transverse or longitudinal direction of the laminated ME composite, the constitutive equation can be directly solved by an averaging method to estimate effective material parameters. The transverse ME coefficient is given as

$$\alpha_{E,31} = \frac{E_3}{H_1} = \frac{-f(1-f)(q_{11}+q_{21})d_{31}}{{}^{p}\varepsilon_{33}\bar{s} - 2fd_{31}^2}. \tag{1}$$

The longitudinal ME coefficient is given as

$$\alpha_{E,33} = \frac{E_3}{H_3} = \frac{2f(1-f)d_{31}q_{31}\mu_0\bar{s}}{\left(2fd_{31}^2 - {}^{p}\varepsilon_{33}\bar{s}\right)\left[\overline{\mu s} + 2q_{31}^2(1-f)^2\right]}. \tag{2}$$

Here, q is the piezomagnetic coupling coefficient, d is the piezoelectric coupling coefficient, ε is the dielectric constant and $\bar{s}$ is the effective compliance. $\bar{\mu} = f\mu_0 + (1-f)^m\mu_{33}$, where μ_0 and μ_{33} are permeability of the free space and the magnetic phase. Superscript p denotes the piezoelectric phase. Here, f is the volume fraction of the magnetic phase:

$$f = \frac{v_{Terfenol-D}}{v_{Terfenol-D} + v_{PZT}} \tag{3}$$

where v is the volume of the PE and PM phases, respectively. The T–P, P–T–P and T–P–T–P samples all have the same material parameters, except for the volume fraction of the magnetic phase. The volume fractions of the three samples are $f_{T-P} = 0.85$, $f_{P-T-P} = 0.66$ and $f_{T-P-T-P} = 0.76$, respectively, based on Eq. (3). The calculation results demonstrated

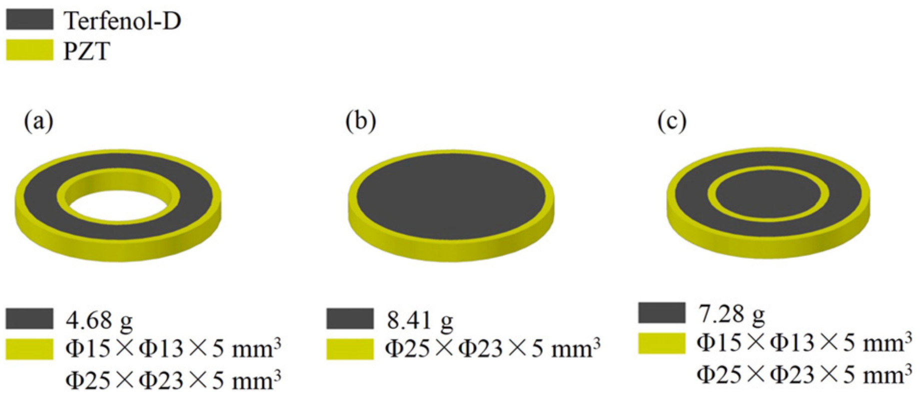

Fig. 1. Schematics illustrating the geometrical arrangement of the Terfenol-D/PZT cylindrical ME composites: (a) P–T–P; (b) T–P; (c) T–P–T–P.

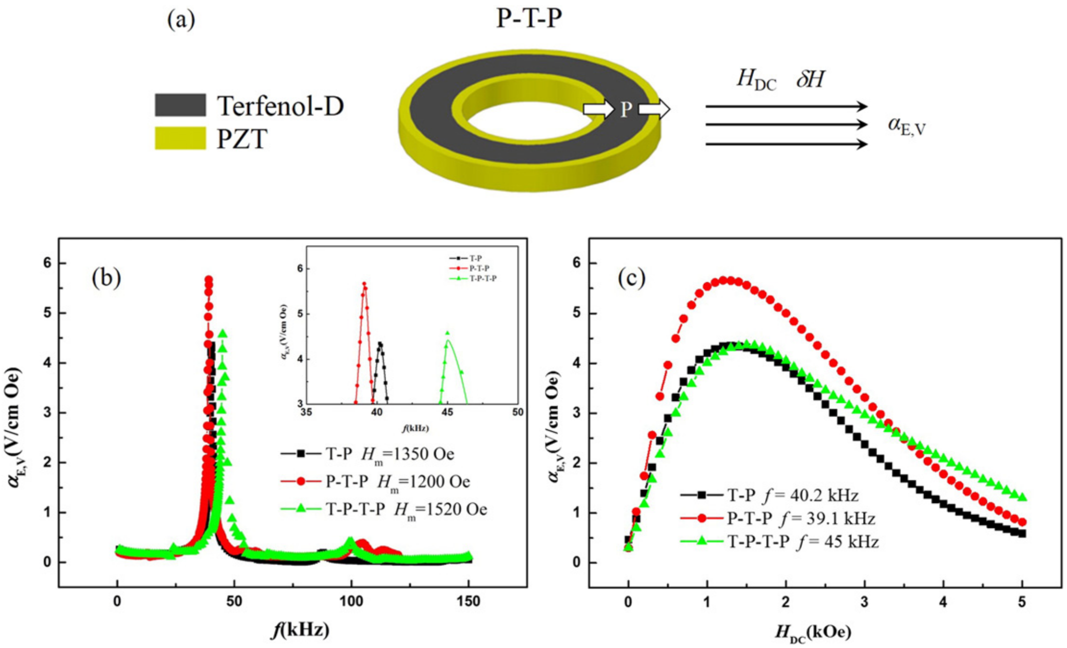

Fig. 2. (a) Schematic of the composite mode (the directions of applied magnetic field and polarization) (showing P–T–P as an example); (b) The $\alpha_{E,V}$ dependence on the AC magnetic field frequency (f) at the optimal magnetic field (H_m) of the three samples, corresponding to the maximum ME coupling; (c) The H_{DC} dependence of $a_{E,A}$ at the resonance frequency.

that the P–T–P composite has higher $a_{E,V}$, which is attributed to an optimal volume fraction (f) of the PM phase, according to the analogous computation results (optimal $f \sim 0.6$) [24]. Additionally, a larger A_{eff} can create more efficient interfacial strain coupling between the PM and the PE phases with the same volume fraction [25]. Namely, when f is constant, the following equation can be used:

$$\alpha_E \propto A_{eff}. \quad (4)$$

The P–T–P composite, which had an optimal volume fraction and the largest A_{eff}, yielded the highest ME voltage coefficient of the three composite samples.

Additionally, the ME voltage coefficient of the P–T–P composite can also be enhanced by the series multi-electrode effect [18]. While the P–T–P and T–P–T–P composites both have series multi-electrode effect, the T–P–T–P composite consists of two parts of positive magnetostrictive Terfenol-D phase. In the magnetic field, the inner and the outer two parts of the magnetostrictive phases, produce normal stress at the same time along the radical direction, from the center of the circle to PZT, inducing PZT dielectric polarization, thus producing the ME effect [23]. In the series mode, the inner and the outer PZT layers contribute to the ME voltage coefficient together [18]. However, for the inner PZT layer of the T–P–T–P composite, normal stress works on both inner and outer surfaces following along the same direction, weakening the polarization [23]. Hence, it weakens the overall ME effect of the T–P–T–P composite.

The measurements of the ME efficiency factor (ME-EF) were conducted. The ME-EF is defined as the α_E contribution of the unit weight PM material, using ME-EF $= \alpha_{E,V}/m_{Terfenol\text{-}D}$. For the P–T–P composite, 5.8 V/cm·Oe is divided by 4.68 g, resulting in the ME-EF of 1.24 V/

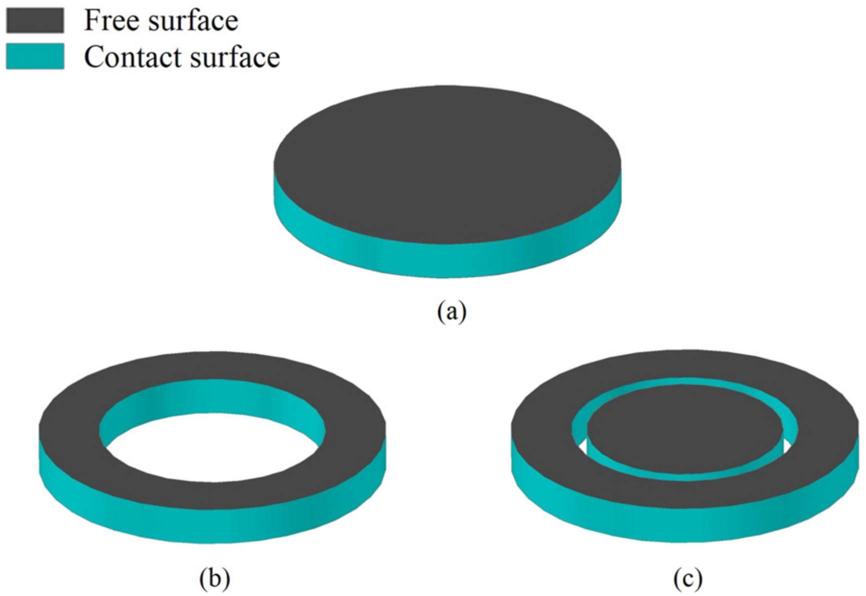

Fig. 3. Schematics illustrating the contact surfaces and the free surfaces of the composites (without PZT): (a) P–T–P; (b) T–P; (c) T–P–T–P.

Table 1
ME voltage coefficients ($\alpha_{E,V}$), ME efficiency factor (ME-EF) and effective working surface specific value (A_{eff}) of the three samples.

Sample	$\alpha_{E,V}$ (V/cm·Oe)	ME-EF (V/cm·Oe·g)	A_{eff} (Contact/free)
T–P	4.4	0.52	0.43
P–T–P	5.8	1.24	1.25
T–P–T–P	4.6	0.63	1.08

cm·Oe·g. It is the highest in the three composites, even higher than the composite with excellent $\alpha_E = 24$ V/cm·Oe (ME-EF = 0.92 V/cm·Oe·g), which consists of 26 g Terfenol-D and four PZT parts connected in series, as reported previously by Zuo [18]. The $\alpha_{E,V}$, ME-EF and A_{eff} of each sample are listed in Table 1.

The outstanding ME-EF of T–P–T cylinder composite represents the T–P–T mode of the ME composite with more efficient utilization of the PM material. It is a result of A_{eff} of the PM and PE phases, boundary and stress conditions of the PM and PE layers and the series multi-electrode effect [18,19]. Hence, the P–T–P composites have outstanding advantages of both high ME voltage coefficients and less usage of the PM material, compared with the T–P and the T–P–T–P composites. The P–T–P layered configuration is just an elementary designed structure, not an optimal design. This study provides a way for efficient utilization of PM materials for the ME composites. Similar, but even better structural designs can widely benefit from cost efficiency and result in lighter magnetic sensors, magnetometers, transducers and stray capacitor devices [26–28].

4. Conclusions

In summary, the study investigated the ME voltage coefficients of the mono/multi-electrode cylinder layered Terfenol-D/PZT ME composites with three different structures for enhancing magnetoelectric efficiency. The ME voltage coefficient, $\alpha_{E,V}$, of the P–T–P mode composite is 5.8 V/cm·Oe at the resonant frequency, and the ME-EF is 1.24 V/cm·Oe·g, which is more than two times higher than the T–P and the T–P–T–P modes. The results of the study demonstrate that this type of hollow multi-electrode cylinder ME composite uses lower amounts of PM materials, but yields higher/better ME voltage coefficients. This finding indicates that creating similar structural designs will not only reduce the weight, but also cost to manufacture ME devices for practical applications, such as magnetic sensors, earth-magnetism navigation, mineral exploration, transducers and magnetometers.

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