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Geometry effects on magnetoelectric performance of layered Ni/PZT composites

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ABSTRACT

Layered magnetoelectric (ME) composite structures of varying geometry consisting of PZT and Ni layers were prepared by electrodeposition. Trilayered plate, bilayered and trilayered cylindrical structures' ME performance was compared. The ME voltage coefficient increased with Ni layer thickness. Cylindrical composites show better ME performance than the plate structures with the same magnetostrictive-piezoelectric phase thickness ratio under high applied magnetic fields at resonant frequencies. Bilayered cylindrical Ni/PZT structure has the best performance in axial mode under high magnetic field, exhibiting linear ME voltage coefficient dependence on the applied magnetic field, which makes it a promising candidate for magnetic field sensor applications. Cutting the ring along the axial direction drastically decreased its performance.

1. Introduction

Layered multiferroic materials are candidates for the next-generation multifunctional devices [1–6]. In these structures, the interaction between ferroelectric and ferromagnetic layers produces new coupled magnetoelectric (ME) effect [7,8]. The ME response appears as an electric polarization upon applying magnetic fields and/or a magnetization upon applying electric fields, and has been observed in some single-phase materials [9,10], including $BiFeO_3$ and $BaMnF_4$ [11,12]. The ME effect comes from the local exchange between the internal orderly magnetic structure and ferroelectric sub-lattice [13]. Unfortunately, single-phase materials exhibit weak ME effect, which explains their limited application. Magnetoelectric layered composite structures provide an alternative, exhibiting higher ME effect due to mechanical coupling between piezoelectric and ferromagnetic layers [14]. When magnetic filed is applied to the ferromagnetic layer, it deforms, transporting mechanical strain onto the piezoelectric layer, thus generating an electric potential. The magnetoelectric coefficient is the voltage generated in the piezoelectric due to the applied magnetic field per piezoelectric thickness, and has the units of V/(cm Oe). Ferroelectric and ferromagnetic layered materials have Curie and Neil temperatures above room temperature, and exhibit larger piezoelectric and piezomagnetic effects compared with traditional single-phase ME materials.

There are two major types of synthesized ME composites: particulate and layered composites. Particulate composites include sintered and organic solidified composites [15–20]. The ME effect is weak in these sintered structures because of the diffusion between phases and the seepage phenomenon. The same problem exists with organic solidified composites made by hot pressing of piezoelectric and piezomagnetic granular materials.

Layering has become a popular method for synthesizing ME composites. This is especially true for joining together piezoelectric and piezomagnetic phases by bonding or hot pressing [21–28]. The ME effect in layered composites is much higher compared with the particulate composites made from the same materials [29]. However, interfacial effects in laminate composites are inevitable, and constrain the improvement and applications of ME laminate composites due to ageing and fatigue.

Liu and co-workers made layered composites with improved ME effect by bonding layers of varying geometry [30]. However, based on the limitation of the gluing method, only simple planar shapes could be produced, including laminated squares and disks [31,32]. Laletin et al. reported the giant ME effect in layered transition metal/PZT samples synthesized by bonding thin disks of PZT and Fe, Co or Ni with an adhesive [33]. As for more complex shapes, such as cylinders, new techniques had to be developed. It is possible to improve the ME layered composite performance by depositing each layer directly, without the use of a bonding agent, which provides much better mechanical coupling and can produce more complex shapes that will help improve the ME effect.

Electrodeposition is widely used for making composite functional materials with good interfacial adhesion. Electrodeposition process has the ability to coat complex shapes, while controlling coating thickness and composition [34–36]. Magnetic materials, including Fe, Co, Ni and their alloys can be deposited by electrodeposition from the corresponding salt solutions. Moreover, the

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bonding glue layer can be avoided with this method. We utilized electrodeposition to make layered ME composites with complex shapes and improved ME properties, which are discussed later.

2. Experimental procedures

Three different structure types were made, and include planar trilayered Ni/PZT/Ni, cylindrical trilayered Ni/PZT/Ni, and cylindrical bilayered Ni/PZT composites, presented schematically in Fig. 1. Commercial PZT-5H ceramic ($Pb(Zr_{0.52}Ti_{0.48})O_3$ supplied by the Institute of Acoustics, Chinese Academy of Sciences) was utilized as the piezoelectric layer and electrodeposited pure nickel (Ni) was utilized as the magnetic layer, which had good piezoelectric properties. The main performance parameters of PZT-5H are shown in Table 1.

Sample preparation consisted of the following steps: first, the PZT was mechanically cut into desired shape, then polarized (vector P shows the polarization direction in Fig. 1), and then bathed in a plating solution to electrodeposit Ni. The thickness of Ni was controlled by the deposition time. For a plate layered ME composite, shown schematically in Fig. 1(a), the dimensions of PZT center layer are $W \times L \times t_{PZT} = 10\,\text{mm} \times 20\,\text{mm} \times 0.25\,\text{mm}$, where t_{PZT} is the thickness of PZT. Varying thickness Ni layers were electro-plated on both sides of the PZT samples. The deposition times were 1, 2 and 4 h, which resulted in 0.1, 0.2 and 0.4 mm thick Ni layers, respectively.

For the cylindrical trilayered ME composite, shown schematically in Fig. 1(b), the dimensions of PZT are $R_1 \times R_2 \times h\,\text{mm}^3$, where $R_1 = 9\,\text{mm}$ is the inner radius, $R_2 = 10\,\text{mm}$ is the outer radius, $h = 8\,\text{mm}$ is the PZT cylinders height and $t_{PZT} = (R_2 - R_1)$. Ni was deposited on both inner and outer PZT cylinder surfaces for 10 h, which resulted in total 1 mm Ni layer thickness (0.5 mm on each side of the cylinder). For the cylindrical bilayered structure, shown schematically in Fig. 1(c), the PZT cylinder height is 3 mm ($h = 3\,\text{mm}$), and the other two dimensions are the same as for the trilayered cylindrical structure ($R_1 = 9\,\text{mm}$ $R_2 = 10\,\text{mm}$). The corresponding PZT radii are R_1 and R_2 shown in Fig. 1(c). Besides having smaller height compared with the trilayered cylindrical structure, Ni was only deposited on the outer PZT cylinder surface for 20 h, resulting in 1 mm Ni layer thickness. For both cylindrical samples, PZT was polarized in the radial directions (P in Fig. 1(b) and (c)).

Ni electrodeposition process is described in detail elsewhere [37]. Nickel aminosulfonate plating solution (concentration of 600 g/L) was used due to its stability, rapid plating speed and small film residual stresses. Nickel chloride (20 g/L) was added to the plating bath to help the anode dissolution. Boric acid (20 g/L) acted as a buffer to stabilize the plating solution pH. The pH value was adjusted to 4 by using sulfamic acid and sodium hydroxide. Surfactant of sodium lauryl sulfate (0.1 g/L) was added to prevent pinholes on the film surface. The total volume of the electro-plating solution was about 1 L, and plating occurred at 60 °C with 5 A/dm^2 current density.

The magnetoelectric measurement system is described in detail elsewhere [38]. The ME voltage coefficient was calculated based on $\alpha_E = \delta V/(t_{PZT} \cdot \delta H)$, where δH is the amplitude of the sinusoidal magnetic field generated by Helmholtz coils. For the plate sample, the polarization direction is along the PZT thickness (transverse direction), denoted as P in Fig. 1(a). We can obtain ME voltage coefficients effect of α_E^{T-L} and α_E^{T-T} when the magnetic fields are applied in the longitudinal and transverse directions, respectively. Magnetoelectric voltage coefficient α_E^{T-L} direction notation used in this paper is the following: the first superscript T denotes the transverse PZT polarization direction and L denotes the longitudinal magnetic applied direction. Similarly, α_E^{T-T} assumes that the PZT thickness is taken along the transverse plate direction, and the magnetic field is applied in the same plate transverse direction.

For these cylindrical layered samples, two ME voltage coefficients α_E^{R-A} and α_E^{R-V} were obtained corresponding to two conditions where H_{DC} and δH were applied along the cylinder axis, or in the vertical direction along its diameter, respectively. For α_E^{R-A}

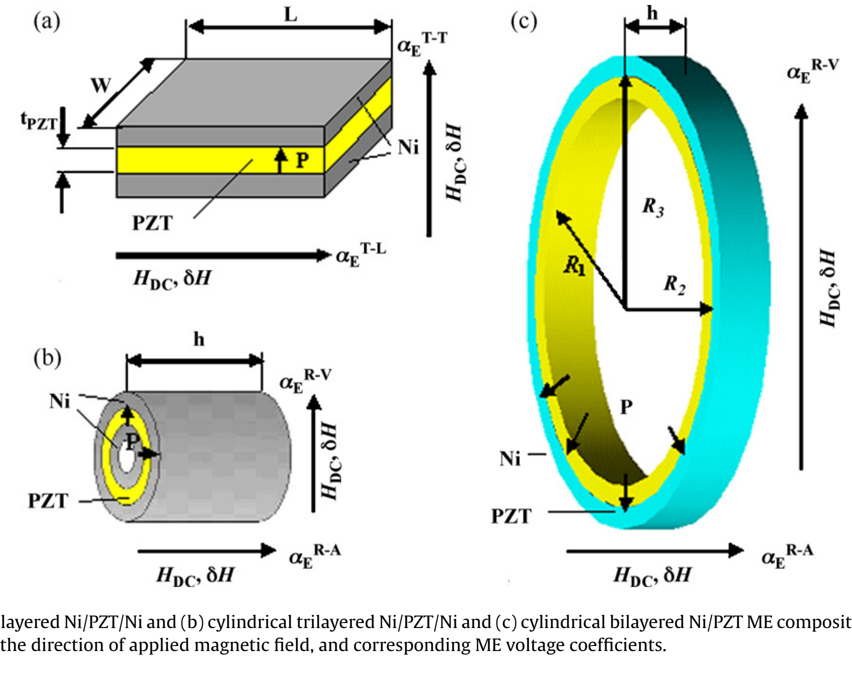

Fig. 1. Schematic of (a) planar trilayered Ni/PZT/Ni and (b) cylindrical trilayered Ni/PZT/Ni and (c) cylindrical bilayered Ni/PZT ME composite. Vector P shows PZT polarization direction. Other vectors identify the direction of applied magnetic field, and corresponding ME voltage coefficients.

Table 1
The main performance parameters of PZT-5H.

d_{33} ($\times 10^{-12}$ C/N)	d_{31} ($\times 10^{-12}$ C/N)	α ($\times 10^{-6}$ °C^{-1})	T_C (°C)	ε	K	tgδ	ρ (kg/m^3)	Q_m
500	–175	10	300	1750	0.65	0.02	7.5×10^3	50