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Magnetoelectric effect in layered composites with arc shape*

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Magnetoelectric (ME) layered Ni/PZT/Ni composites with arc shape have been prepared by using electroless deposition. The ME effect is measured by applying both constant and alternating magnetic fields in longitudinal and transverse directions. The longitudinal ME voltage coefficient is much larger than the transverse one. With the increase of arc length or decrease of curvature, the resonance frequency of layered arc Ni/PZT/Ni composites gradually decreases, while the maximum of the ME voltage coefficient of the composites increases monotonously. The influence of the arc length and the curvature on ME coupling is discussed. The flat interface between the ferromagnetic and the piezoelectric phases in layered ME composites is believed to provide large ME voltage coefficient.

Keywords: magnetoelectric, arc-shaped, electroless deposition, resonance frequency

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1. Introduction

Magnetoelectric (ME) materials, which can display an electric polarization induced by an applied magnetic field or conversely a magnetization induced by an applied electric field, have recently stimulated tremendous fundamental and practical interests for use as sensors, actuators and transducers.[1−3] The ME single phase materials are difficult to apply in practical applications due to their weak ME response and low Curie or Néel temperature.[4−6] The ME effect is rather strong in composite systems in comparison with single phase materials.[7,8] The absence of a substantial leakage current in layered composites, coupled with ease of polarization, leads to an enhancement of piezoelectric and ME effects. A magnetic field applied to the composite will induce strain in the magnetostrictive layer which is passed along to the piezoelectric layer, where it induces an electric polarization.[9]

To obtain desirable ME properties, layered ME composites, with Ni, Fe–Co alloys, or rare-earth alloys as the magnetostrictive phase and with lead zirconate titanate (PZT), lead magnesium niobate-lead titanate (PMN-PT) as the piezoelectric phase, have been made into different structures.[10−19] Wan *et al.*,[20] who first systematically studied the ME properties in arc-shaped Terfenol-D/epoxy-PZT bilayer, they obtained a resonance ME voltage coefficient of 14.6 $\mathrm{V\cdot cm^{-1}\cdot Oe^{-1}}$ (1 Oe = 79.5775 A/m) at frequency of 12.5 kHz. The ME effect in layered ME composites is a product property of the magnetostrictive and piezoelectric effects. The coupling between magnetostrictive and piezoelectric layers is strongly influenced by its structure. Therefore the arc length and curvature may influence the ME coupling of layered arc ME composites. However, this has rarely been reported so far. Electroless deposition is widely used to obtain functional films with good interfacial bonding on various substrates, even nonmetallic materials.[21] It provides a convenient method to fabricate ME layered composites with complex shapes and good mechanical coupling at interfaces with neither electrodes nor bonding layers. In this work, we studied ME coupling in layered arc Ni/PZT/Ni composites and investigated the influence of arc length and curvature on ME properties.

2. Experimental details

Experiments presented here are based on layered arc Ni/PZT/Ni composites with commercial PZT as the piezoelectric phase and electroless deposited Ni as the magnetostrictive phase. The PZT ceramics were cut into arc pieces with thickness of 0.4 mm and width of 5 mm. The pretreatment process applied

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prior to the electroless deposition consisted of roughening, sensitization, activation and reduction. The PZT samples were dipped into a nickel sulfate bath for the deposition of Ni layers. The bath composition and operation parameters of electroless deposition are described in detail elsewhere.[22,23] The pH of the bath was adjusted by using sodium hydroxide and measured with an electronic pH meter (Orion model 720A). The thickness of Ni layers was controlled by the electroless deposition time. In this experiment, the resulting total thickness of the two Ni layers was approximately 100 μm and identical for all the composites. After the electroless deposition, layered arc Ni/PZT/Ni composites were poled in an electrical field of 30 kV·cm^{-1}–50 kV·cm^{-1} applied perpendicularly to the sample plane. The geometric arrangement of layered arc Ni/PZT/Ni composites is shown illustratively in Fig. 1. The R_1 is the inner arc radius of PZT and R_2 is the outer arc radius of PZT. The $R = (R_1 - R_2)/2$ is the average radius of layered arc Ni/PZT/Ni composites.

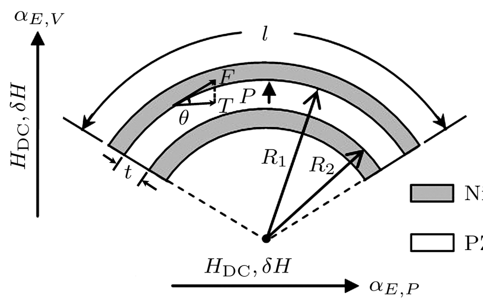

Fig. 1. Schematic geometric arrangement of the layered arc ME composites.

The structure of Ni electroless deposited onto PZT layer was characterized by X-ray diffraction (XRD, D8 Advance Bruker) with Cu K-α radiation. The cross section of layered arc Ni/PZT/Ni composites was investigated by using scanning electron microscopy (SEM, LEO-1530VP). The ME effect of layered arc Ni/PZT/Ni composites was measured by applying both constant (H_{DC}) and alternating (δH) magnetic fields in longitudinal and transverse directions. The induced voltage signal δV across the sample was amplified and measured by an oscilloscope. The DC bias magnetic field H_{DC} could be changed in the range –8000 Oe–8000 Oe and the superimposed AC magnetic field δH was generated by Helmholtz coils. The ME voltage coefficient was calculated based on $\alpha_E = \delta V/(t_{\mathrm{PZT}}\delta H)$, where t_{PZT} is the thickness of PZT layer. In this experiment, $\delta H = 1.2$ Oe as the amplitude of the AC current through the coil is equal to 1 A.

3. Results and discussion

The typical XRD pattern of Ni electroless deposited onto PZT layer with pH = 10.7 and bath temperature $T = 363$ K is shown in Fig. 2(a). It can be seen that all peaks can be identified. There are no additional phase peaks apart from FCC Ni. The cross-sectional SEM images of layered arc Ni/PZT/Ni composites are shown in Fig. 2(b). It reveals that the Ni layer is in direct contact with the PZT layer without interlayer. This indicates that there is a good interface coupling between Ni and PZT layers. The XRD patterns of the deposited Ni layers and cross-sectional SEM images of layered arc Ni/PZT/Ni composites for all the samples are the same due to same operation parameters in electroless deposition process.

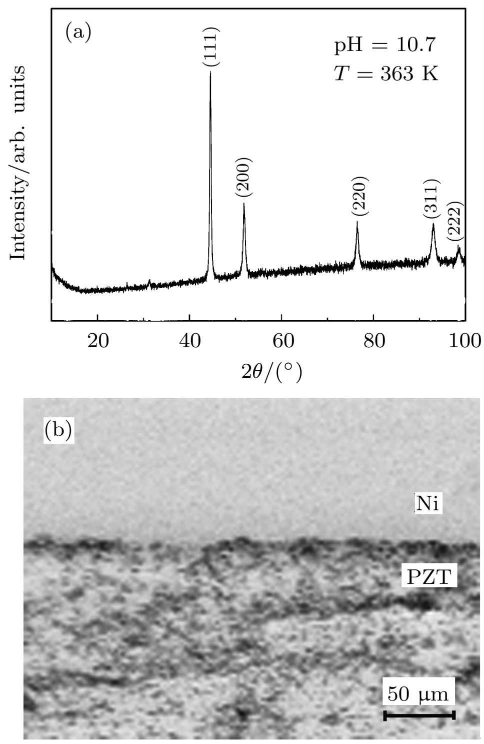

Fig. 2. (a) The XRD pattern of the deposited Ni layer with pH = 10.7 and T = 363 K. (b) The cross-sectional SEM images of layered arc Ni/PZT/Ni composites showing the Ni–PZT interface.

Figure 3(a) shows representative data on bias magnetic field H_{DC} dependence of ME voltage coefficient (α_E). The magnetic fields are applied in longitudinal and transverse directions (shown in Fig. 1), $\alpha_{E,P}$ and $\alpha_{E,V}$ are corresponding ME voltage coefficients respectively. The measurements were carried out at $f = 1$ kHz on a layered arc Ni/PZT/Ni composites with the dimension of 28 mm (length, l) ×5 mm (width, w)×0.41 mm (thickness, d). Data are shown for increasing H_{DC}. When H_{DC} increases from zero, it is seen that α_E depends strongly on H_{DC}. One observes a sharp increase in $\alpha_{E,P}$ to a maximum at $H_{\mathrm{m}} = 200$ Oe and $\alpha_{E,V}$ to a maximum at $H_{\mathrm{m}} = 300$ Oe. With further increase of H_{DC}, $\alpha_{E,P}$ and $\alpha_{E,V}$ decrease rapidly to a minimum. The maximum value of $\alpha_{E,P}$ is larger than that of $\alpha_{E,V}$. The measurements on the AC magnetic field frequency f dependence on the ME coupling are also performed. For such studies the bias magnetic field H_{DC} was set at H_{m} and α_E was measured as f was varied from 1 kHz to 150 kHz. Typical α_E versus f profile is shown in Fig. 3(b). It can be seen that no remarkable frequency dispersion is observed except for the resonance region. When f increases, we observe a sharp peak in $\alpha_{E,P}$ and $\alpha_{E,V}$ at resonance frequency of $f_{\mathrm{r}} = 70.6$ kHz. The maximum value of $\alpha_{E,P}$ for $H_{\mathrm{DC}} = 200$ Oe is much larger than that of $\alpha_{E,V}$ for $H_{\mathrm{DC}} = 300$ Oe. Pan *et al.* studied the shape demagnetization effect on ME voltage coefficient of layered ME composites and obtained an expression[24]

$$\alpha_E \propto (1+\beta)/N, \tag{1}$$

where β is magnetostriction factor for a given shape material, N is the demagnetization factor in the direction of applied magnetic fields. The demagnetization factor with magnetic fields applied in longitudinal direction is much smaller than that with magnetic fields applied in transverse direction, so the longitudinal $\alpha_{E,P}$ is much larger than the transverse $\alpha_{E,V}$.

The resonance frequency of the transverse vibration mode for layered Ni/PZT/Ni composites is given by[20]

$$f_{\mathrm{r}} = \frac{1}{2l}\sqrt{\frac{1}{\overline{\rho}\ \overline{s_{11}}}}, \tag{2}$$

where f_{r} is the resonance frequency, l is the length of layered Ni/PZT/Ni composites, $\overline{\rho}$ is the average density of the composites, it is determined by $\overline{\rho} = \nu_m\rho_m + \nu_p\rho_p$, $\overline{s_{11}}$ is the equivalent elastic compliance and given by $\overline{s_{11}} = {}^m s_{11}{}^p s_{11}/(v_m{}^p s_{11} + v_p{}^m s_{11})$, where v_m and v_p are the volume fractions of Ni and PZT layers, respectively, ${}^m s_{11}$ and ${}^p s_{11}$ are the respective elastic compliances of the layers.

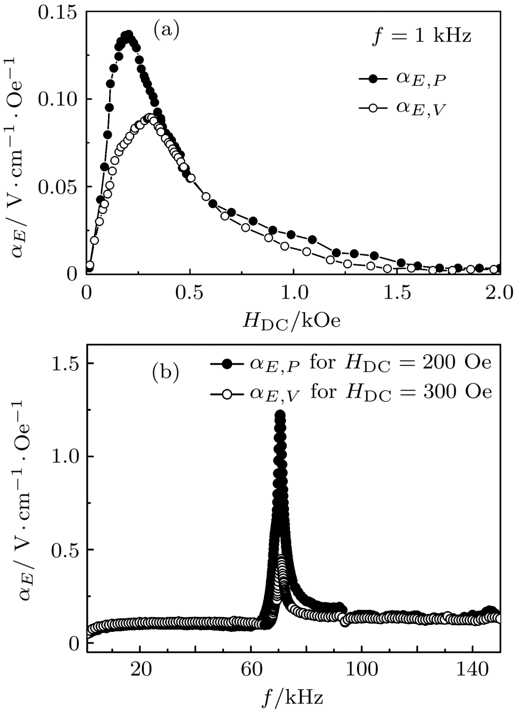

Fig. 3. (a) Bias magnetic field H_{DC} dependence of ME voltage coefficient ($\alpha_{E,P}$ and $\alpha_{E,V}$) at $f = 1$ kHz. (b) The AC magnetic field frequency f dependence on $\alpha_{E,P}$ at $H_{\mathrm{DC}} = 200$ Oe and $\alpha_{E,V}$ at $H_{\mathrm{DC}} = 300$ Oe for layered arc Ni/PZT/Ni composites with the dimension of 28 mm×5 mm×0.41 mm.

From the above analyses, one can see that the resonance frequency is inversely proportional to the arc length l for given curvature. The dependence of the resonance frequency f_{r} on arc length l is shown in Fig. 4. It can be seen that the resonance frequency decreases as the arc length increases, which is in good agreement with the theoretical analysis. The dependence of the maximum ME voltage coefficient $\alpha_{E,P}$ on arc length l is also shown in Fig. 4. With the increase of arc length from 28 mm to 42 mm the maximum $\alpha_{E,P}$ of layered arc Ni/PZT/Ni composites with an arc radius of 11 mm gradually increases from 0.83 V·cm^{-1}·Oe^{-1} to 1.37 V·cm^{-1}·Oe^{-1}. The resonance frequency and the maximum ME voltage coefficient are strongly influenced by arc length. These results indicate that one can obtain a higher ME voltage coefficient and lower resonance frequency by extending arc length.

Figure 5 shows representative data on arc radius R dependence of the resonance frequency f_{r} and the maximum ME voltage coefficient $\alpha_{E,P}$. The arc length l of layered Ni/PZT/Ni composites is a constant of 28 mm. With the increase of arc radius (decrease of curvature) from 11 mm to 13 mm the resonance frequency of layered arc Ni/PZT/Ni composites gradually decreases from 75.9 kHz to 66.3 kHz, while the maximum ME voltage coefficient $\alpha_{E,P}$ of the composites increases from 0.83 $V \cdot cm^{-1} \cdot Oe^{-1}$ to 1.76 $V \cdot cm^{-1} \cdot Oe^{-1}$. This suggests that the resonance frequency and the maximum ME voltage coefficient are also influenced by the curvature of layered arc Ni/PZT/Ni composites.

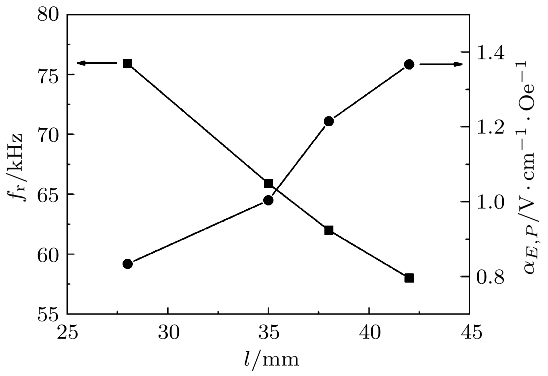

Fig. 4. The dependence of the resonance frequency f_r and the maximum ME voltage coefficient $\alpha_{E,P}$ on arc length l with an arc radius of 11 mm.

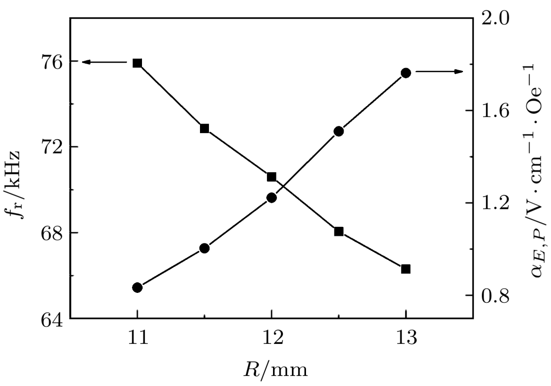

Fig. 5. The dependence of the resonance frequency f_r and the maximum ME voltage coefficient $\alpha_{E,P}$ on arc radius R for layered arc Ni/PZT/Ni composites with an arc length $l = 28$ mm.

In composite materials the ME effect is generated as a coupling of magnetic–mechanical–dielectric behaviour. The stress passed along to the piezoelectric layer from the magnetostrictive layer plays an important role in ME coupling. As shown in Fig. 1, F is the stress induced by magnetostrictive effect in Ni layer, T is the stress passed along to the PZT layer. The $T = F{\cdot}\cos\theta$, θ is the angle between T and F. With the increase of arc radius (decreasing in curvature), θ decreases. When F is constant, T increases as θ decreases. This leads to an enhancement in ME coupling. The fact that the maximum ME voltage coefficient $\alpha_{E,P}$ of the composites increases with the curvature implies that the flat interface between the ferromagnetic and the piezoelectric phases in layered ME composites may provide large ME voltage coefficient.

One should note that the dependence of ME properties on the arc length and curvature cannot be attributed to the structural factors and there is no structural difference observed on all the samples. From XRD patterns it can be seen that all the deposited Ni layers have similar fcc structure and there is only Ni phase in all the Ni layers. The cross-sectional SEM shows that the electroless deposited Ni layers contacts well with PZT layers for all the samples.

4. Conclusions

The ME effect of layered arc Ni/PZT/Ni composites have been studied. The ME voltage coefficient with magnetic fields applied in longitudinal direction is much larger than that with magnetic fields applied in transverse direction. Both arc length and curvature influence the ME coupling of layered arc Ni/PZT/Ni composites. The resonance frequency of layered arc Ni/PZT/Ni composites gradually decreases with the increase of arc length or arc radius (decreasing in curvature). But the maximum ME voltage coefficient $\alpha_{E,P}$ of the composites increases as arc length or arc radius increases. Thus, one can obtain a higher ME voltage coefficient and lower resonance frequency by increasing arc length or decreasing curvature. Large ME voltage could be expected in layered composites with flat ferromagnetic–piezoelectric interface.

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