Effect of Ni and FeCoV films on magnetoelectricity of piezoelectric Pb(Mg1/3Nb2/3)O3–PbTiO3 sandwich structures

Zhaofu Du a, Sam Zhang b,*, Lei Wang a, Dongliang Zhao a

a Research Institute of Functional Materials, Central Iron and Steel Research Institute, Beijing 100081, China
b School of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

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A B S T R A C T
To improve the magnetoelectric (ME) property of piezoelectric thin film systems, Ni and FeCoV magnetic films are respectively used to sandwich piezoelectric foil Pb(Mg1/3Nb2/3)O3–PbTiO3 (PMN–PT) in the middle. The ME voltages and coefficients have been characterized in the longitudinally magnetized and transversely polarized sandwich structures. The measurement was conducted under a static magnetic field superimposed with an alternating magnetic field. The influences of the static and the alternating field strength are discussed. The peak ME coefficient obtained in the FeCoV film-sandwiched structure reached 27.4 × 10^{-3} mV/cmA/m, which is about five times greater than the Ni film-sandwiched counterpart. A linear relationship is observed between the magnetoelectric voltage and the alternating field strength under a static field of 31.8 kA/m for both structures. As FeCoV is also oxidation-resistant and low in cost, the FeCoV film-sandwiched structure (i.e., FeCoV/PMN–PT/FeCoV) makes a good candidate in miniature ME device applications.

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1. Introduction
Magnetoelectric (ME) materials have recently stimulated much fundamental and application research interest due to potential applications in multifunctional devices [1,2]. However, the Néel temperature of the single-phase magnetoelectric materials is usually under 130 K thus it is unable to use for practical application at room temperature. For room temperature applications, ferroelectric and ferromagnetic composites are designed and their massive applications have been realized in multifunctional devices such as magnetoelectric sensors, actuators and transducers [3,4].

Giant magnetostrictive and excellent piezoelectric materials (such as Tb1–xDyFe2–y (Terfenol-D) and Pb(Mg1/3Nb2/3)O3–PbTiO3 (PMN–PT)) have good ME response [5–7]. Though ME coefficient of 58.8 × 10^{-3} V/cmA/m has been obtained for various laminated PMN–PT/Terfenol-D composite structures, Terfenol-D is expensive and easily oxidized, thus this composite structure encounters difficulties in practical applications. In addition, bulk material lamination is not suitable in miniaturized devices. Thin film ME structures are thus attempted such as NiFe2O4 film/BaTiO3 film [8] and TbFe film/Pb(Zr,Ti)O3 film/TbFe film [9]. But NiFe2O4 has low magnetostrictive property and TbFe needs a large magnetic field, so both of them have difficulties in practical devices.

To make ME coefficient α_{ME} measurable, consider the following expression [10]:

\[ \alpha_{ME} = \frac{\delta E}{\delta H} = \left( \frac{\delta V_i}{\delta t} / V_i \right) / \delta H \] (1)

where, \( H \) is the applied magnetic field strength, \( E \) and \( V_i \) are the electric field strength and voltage induced across the piezoelectric layer, and \( r \) is the thickness of the sample or more accurately the effective distance between the electrodes of the piezoelectric phase. From Eq. (1), \( \alpha_{ME} \) can be improved by decreasing \( \delta H \) at constant \( \delta V_i \). As soft magnetic materials, such as FeCoV and Ni, are sensitive to magnetic field, they can be considered for use in ME structures.

In this paper, magnetron sputtered Ni and FeCoV films were used to sandwich a piezoelectric PMN–PT foil for measurement of the ME effect.

2. Experimental details
Commercially obtained high grade piezoelectric material, i.e., PMN–PT in the form of a foil of 0.2 mm in thickness, is used as the core of the sandwich. A 0.2 mm thick foil provides enough mechanical strength thus renders additional substrate unnecessary. The foil, polarized in its thickness direction, was cut into rectangular shape of 20 × 10 mm. Magnetron sputtering was used to deposit Ni and FeCoV thin films respectively on both sides of the foil to form sandwich structures of Ni/PMN–PT/Ni (or denoted as NPN from now on) and FeCoV/PMN–PT/FeCoV (or denoted as FPFP from now on) as sketched in Fig. 1. After reaching a base pressure of 3.5 × 10^{-4} Pa, the chamber was backfilled with Ar to a pressure of 1.0 Pa. At a target power density of 300 V (DC) × 80 mA/(35 mm)^2, or 0.6 W/cm^2, a 4-h sputtering of Ni target gives rise to 3.62 μm of Ni film and 4-h sputtering of FeCoV target gives rise to 2.80 μm of Fe_{49.5}Co_{49}V_{1.5} film. As the magnetstriction response for Ni or FeCoV films is unavailable, Ni and FeCoV rectangular plates of 20 × 10 × 0.4 mm were prepared by grinding from 1 mm thick Ni and FeCoV plates, and
their linear magnetostriction measured using a standard strain-gauge technique. The sputtered films were analyzed by X-ray diffraction (XRD) [D/Max-RB, Rigaku] with Cu Kα radiation with step of 0.02° in 30–80° (2θ) and Scanning Electron Microscopy (SEM) [XL30S-FEG, Philips] with incident electron energy of 15 kV at room temperature.

The ME effect of the sandwich structures was measured under constant magnetic field strength (H_{OC}) superimposed with a small alternating field strength (H_{AC}) both applied parallel to the sample surface in the longitudinal direction. The induced ME voltage is in the thickness direction. As a small alternating magnetic field is superimposed on the static field, the measurement is “dynamic” [12] as illustrated in Fig. 2. The static magnetic field H_{OC} was controlled by a DC source [GP-HP-L/±65A] variable from 0 to 318.4 kA/m, and the alternating field H_{AC} variable from 23.4 A/m to 199 A/m was generated by a home-made solenoid supplied by a signal generator [CA1640-02, CALTEK] and monitored by an AC induction magnetometer [CGG-1000]. The magnetic field strength was measured by Tesla meter [T-6 TYPE, ISAS]. The electric signal produced by the sample was measured through a lock-in-amplifier [LI 5640].

3. Results and discussion

3.1. Microstructure of the magnetic films

Fig. 3 plots the XRD patterns of sputtered Ni and FeCoV thin films. Grazing-incidence diffraction at 1° was used to eliminate diffraction from the underneath PMN–PT foil. Crystalline face-centered cubic Ni film is confirmed. In FeCoV film, one peak at (1 1 0) is observed. SEM cross-section and the surface of the magnetic thin films before and after the ME measurement are shown in Fig. 4. Both films are of columnar structures of 50–100 nm in diameter. Before the ME measurement, both films are of smooth surface and no surface cracks (see inset in Fig. 4). Both surfaces developed surface cracks due to magnetic straining. All ME responses reported are measured after the system is stabilized (formation of surface cracks completes).

3.2. Magnetostriction of the magnetic plate

Fig. 5 illustrated the magnetostrictive coefficient (λ) vs. magnetic field (H) of the Ni and FeCoV plates. For Ni, with increasing H, the magnetostrictive coefficient quickly decreased to saturation of about –40 ppm at 79.6 kA/m. In FeCoV, the magnetostrictive coefficient increased sharply to saturation of about 95 ppm at 59.7 kA/m.

3.3. ME effect of the sandwich structures

Fig. 6 illustrated the ME coefficient as a function of static magnetic field strength for both NPN and FPF structures. As seen in Fig. 6, \( \alpha_{ME} \) is strongly dependent on the static field strength and peaks at 35.8 kA/m in NPN with peak value of \( 5.5 \times 10^{-3} \) mV/cm-A/m and at 27.9 kA/m in FPF with peak value of \( 27.4 \times 10^{-3} \) mV/cm-A/m. The saturation magnetic field strengths for both film-sandwiched structures (144.3 kA/m in FPF and 159.2 kA/m in NPN) are greater than that in the magnetic plate (59.7 kA/m and 79.6 kA/m, Fig. 5). This increase in required magnetic field strength is believed to come from overcoming the pinning effect from the underneath piezoelectric foil and the interfacial anisotropy [13]. Fig. 7 plots induced ME voltages over alternating field strengths. A perfect linear relationship is observed under static field of 31.8 kA/m (as ME coefficient peaks at 27.9 kA/m in FeCoV and 35.8 kA/m in Ni film-sandwiched structure, 31.8 kA/m was chosen to test the response to varying alternating fields). As is seen in Fig. 7, FPF has a maximum voltage response about five times that of NPN, corresponding the same difference in ME coefficients depicted in Fig. 6. This makes FPF a perfect candidate in AC magnetic sensing. The large ME voltage and \( \alpha_{ME} \) in FPF come from the large magnetostriction of FeCoV, as explained below.

According to Livingston’s model of coherent rotation of magnetization, the strain generated from magnetostriction under the small amplitudes of alternating magnetic field strength \( H_{AC} \), \( \Delta \varepsilon \) is expressed in [14]:

\[
\Delta \varepsilon = \frac{3\varepsilon_s}{2} \left( \frac{H_{AC}}{H_0} \right)
\]

where, \( \varepsilon_s \) is the saturation magnetostriction constant, and \( H_0 \) is the magnetic anisotropy field. As FeCoV has much larger saturation
magnetostriction than Ni (c.f., Fig. 5), the corresponding strain $\Delta \varepsilon_0$ will be much larger, so will be the resultant stress, as can be seen clearly through [15]:

$$\sigma_p^E = \frac{2E_p t_p \Delta \varepsilon_0}{(1-v)\left(2E_m t_m + E_p t_p\right)},$$

where $E$, $t$, $\Delta \varepsilon_0$, and $v$ are the elastic modulus, thickness, linear strain of the magnetic phases, and Poisson’s ratio, respectively. The subscript $m$ and $p$ represents magnetic phases and the piezoelectric phase, respectively.

For PMN–PT, the induced strain is approximately proportional to the square of the electric field [16]. However, as the $H_{AC}$ is very small (less than 199 A/m), the strain induced by $H_{AC}$ is also very small, the resultant voltage of the PMN–PT is very small, thus $H_{AC}$ is overlaps with the $H_{DC}$ (about 3.18 kA/m). Within this range, we assume that the ME voltage is proportional to the stress, as seen in Fig. 7. As ME voltage and $\alpha_{ME}$ is directly proportional to the stress [15],

$$V_i = 2g_{31} \times t \times \sigma_p^E, \quad (4)$$

$$\alpha_{ME} = \frac{V_i}{t \times H_{AC}} = \frac{2g_{31} \times \sigma_p^E}{H_{AC}} (V/cm\cdot A/m), \quad (5)$$

where $g_{31}$ is the piezoelectric voltage constant. Greater stress ($\sigma$) in FPF therefore gives rise to greater ME voltage ($V$) and ME coefficient ($\alpha$) in FPF structure.

The effect of the magnetic film thickness can also be seen from Eq. (3): the film thickness $t$ appears in both nominator and in the denominator. However, in the denominator, as the piezoelectric foil (in this case, PMN–PT foil) thickness is in a few orders of magnitude larger than that of the magnetic film, the film thickness in the
denominator is neglected (200 μm PMN–PT as compared to a few μm magnetic film), thus stress $\sigma_p$ is directly proportional to the magnetic film thickness $t_m$. In this study, FeCoV film is 2.80 μm while the Ni film is 3.62 μm, should the FeCoV film have the same thickness as Ni film, the ME response would be even better.

4. Conclusions

Magnetron sputtered Ni and FeCoV magnetic films are used to sandwich PMN–PT foil for magnetoelectricity. FeCoV film based sandwich structure results in a maximum magnetoelectric coefficient of $27.4 \times 10^{-3}$ mV/cm•A/m in contrast with that of Ni film counterpart of $5.53 \times 10^{-3}$ mV/cm•A/m (a difference of five times). Similarly the magnetoelectric voltage induced in FeCoV film-based structure is five times more than that induced in the Ni film-based structure. A perfect linear relationship is obtained between the induced magnetoelectric voltage and alternating magnetic field strength. This property may be used for sensing applications.

References