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Magnetoelectric properties of Ni/PZT/Ni layered composite for low field applications

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Abstract A composite material when placed under the external magnetic/electric fields exhibits voltage/induced magnetization is known as magnetoelectric (ME) composite. Such composite materials should have ferroelectric and ferro/ferri magnetic phases as constituents. The magnetoelectric output is exhibited as a product property. Magnetoelectric composites are being used for variety of applications including resonators, filters, phase shifters, optical isolators, actuators and magnetic field sensors. Metal/ferroelectric/metal magnetoelectric composite using Ni and PZT as constituent phases has been fabricated in 2-2 composite pattern to study its product property. The paper presents magnetoelectric studies of Ni/PZT/Ni composite using low dc magnetic field magnetoelectric set-up. Using this ME set-up ME output of Ni/PZT/Ni composite is studied as a function of dc magnetic field. The results were analyzed to identify the useful magnetic field (dc and ac) range in which Ni/PZT/Ni sensor can be utilized for applications.

Introduction

Magnetoelectric effect is defined as exhibition of electrical polarization in presence of external magnetic field or magnetization in presence of external electric field. Many materials including single phase and composites have been reported to exhibit magnetoelectric effect [1, 2]. In case of single phase materials, there exists simultaneous ferroelectric and ferro/ferrimagnetic ordering, which leads to magnetoelectric effect [3]. While, in composites of ferro/piezoelectric and ferro/ferri/piezo magnetic phases, ME output is observed as a product property of two phases, which is absent in either of the phases. The deformation of piezomagnetic/ferrite phase causes the polarization of piezoelectric particles of the composites material, and on the other hand, the electrical polarization of the piezoelectric material causes the change in the magnetization of the piezomagnetic/ferrite phase due to the mechanical coupling of the piezomagnetic and piezoelectric phase.

In recent years, there has been renewed interest in the area of magnetoelectrics due to their wide applications in the field of sensors and actuators [4–6]. However, due to their low magnetoelectric output, single phase materials are not widely used for applications. In turn, composites were observed to show high ME outputs useful for applications [7]. In order to further improve the output, various researchers attempted fabrication of composites with different constituent phases [8–12], and it is well reported in latest reviews [13, 14]. We have previously successfully fabricated metal/PZT/metal composites in 2-2 connectivity for the first time, which exhibited improved ME output [15]. Soon after, Latetsin et al. [16] fabricated bilayers and trilayers with permendur, a ferromagnetic alloy (having magnetostriction 70 ppm) and ferroelectric lead zirconate titanate (PZT) and reported magnetoelectric properties

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under the resonant frequency. Recently, Zhang et al. [17] attempted the single field driven ME effect in highly magnetostrictive rare earth alloy, $Tb_{1-x}Dy_xFe_{2-y}$, and ferroelectric PZT bilayers. The present article is aimed at studying magnetoelectric output of Ni/PZT/Ni composite system at low dc bias fields, in order to ascertain the useful magnetic field range of this composite for potential device applications.

Experimental

Sample fabrication

In the present study, Ni was chosen as the magnetic phase. The desired dimensions of Ni specimens were cut from spec pure (99.9+%) metals obtained from M/s Chempure, USA. The piezoelectric phase chosen for the studies was PZT, which is known to show high-piezoelectric constants, to prepare the laminate composite. The PZT discs were cut from the samples obtained from M/s Concord Electroceramics Ltd, New Delhi, India.

In the present study, PZT pellets were coated with silver paint on the both large surfaces to ensure good ohmic contact. In electric poling process, the PZT sample was heated to 100 °C in an external electrical field of 30 kV/cm, kept at this temperature for about 30 min, and then cooled to room temperature in the presence of the field at the rate of 10 °C/min. Proper electric poling is necessary to obtain magnetoelectric output in composites.

Lead zirconate titanate pellets were machined to thicknesses of 1 mm and a diameter of 5 mm. Ni discs were machined to dimensions of Φ 5 mm × 1 mm. To construct the Ni/PZT/Ni laminate composite, a PZT disc was bonded between two Ni discs along the thickness direction using a silver epoxy adhesive at 80 °C cured for 1 h and 100 °C for half-an-hour to ensure good mechanical coupling between the discs. To ensure proper contact between two phases, external load was applied on the laminates during curing. The electrical wires were connected to the outer surfaces of both the Ni discs to form the output terminals.

Magnetoelectric measurements

In order to realize ME effect, the composite have to be poled both electrically as well as magnetically [8]. For magnetic poling, a sufficiently high-magnetic field (5–10 kOe) is applied to the composite sample such that domains with in the individual crystallites are reoriented and more or less aligned in the direction of the magnetic field. In general, samples are magnetically poled in the same direction as that of the electrical poling. The sample under test (Ni/PZT/Ni laminate) was kept between the pole pieces of the electromagnet magnetically poled in a dc field of 6 kOe for 60 min. A proper poling strategy would ensure magnetic domains to be oriented in the direction of the field. This is necessary for realizing higher magnetoelectric output in the composites.

Magnetoelectric measurements were preformed adopting the dynamic method. In the dynamic method, the samples were kept between the pole pieces of a dc magnet, which can generate the dc magnetic field up to 5 kOe. The ME output was recorded at fixed ac magnetic fields of 2–64 Oe ($f = 1.008$ kHz), superimposed on a varying dc field (in the range of 0.7–5 kOe).

Design and fabrication of ME set-up for low fields

Figure 1 shows a schematic of the testing apparatus. The sample is magnetically loaded perpendicular to the

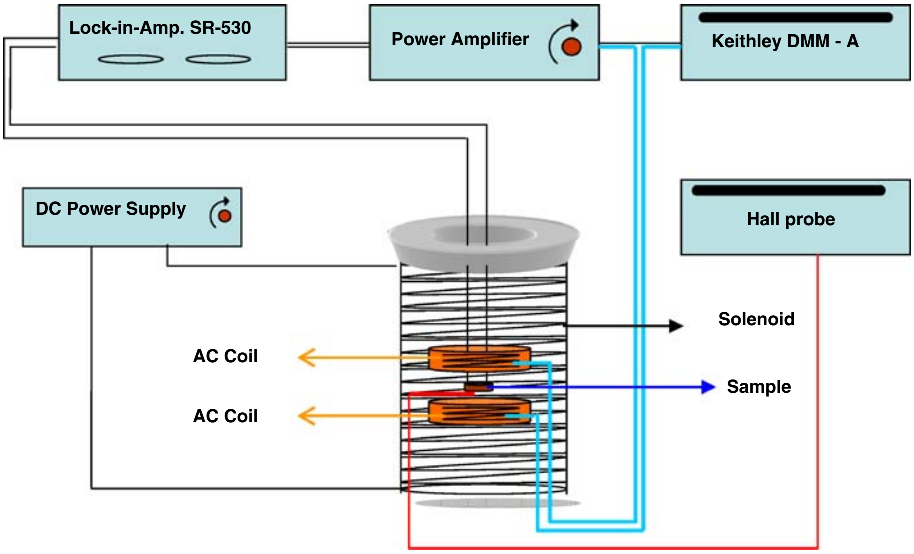

Fig. 1 The schematic of ME set-up for low fields

solenoid having dimensions of 7.56 cm inner diameter, 15.28 cm outer diameter, a length of 30 cm and 3,600 turns. The magnetic field strength produced by dc current through the coil at the centre is measured using an axial hall probe of Digital Gauss Meter (Model No 202).The solenoid produces a dc magnetic field of 0–500 Oe in positive direction and −500 Oe in negative field. The ac Helmholtz coils each having 32 turns are mounted on bakelite cylinder. This cylinder is inserted inside the solenoid along its axis such that the Helmholtz coils are placed at its centre. The sample is loaded at the centre of Helmholtz coils normal to the field. These coils are connected to the Lock-in-Amplifier (Stanford SR-530) through the power amplifier (TZA-4000) as shown in block diagram. Magnetic testing was performed under various dc magnetic biases with a superposed ac magnetic field of 1.008 kHz frequency.

The magnetic field strength *H*, produced by ac Helmholtz coil pair is calculated by the formula $H \approx 0.899NI/R$, where H is the magnetic field in Oersteds, N the total number of the turns per coil, I the current through the Helmholtz coil and R the radius in cm. The current through the Helmholtz coils is measured using Keithley 196 DMM. The experimental set-up was calibrated using theoretically calculated magnetic field values and experimentally observed values calculated using search coil method [18].

Results and discussion

Figure 2 shows the ME output (E) versus dc magnetic field for Ni/PZT/Ni sample. It is observed that with increase in dc bias field, the ME output is found to decrease. For the present Ni/PZT/Ni sample, the output recorded was 173.5 mV/cm at 0.7 kOe dc bias field, and fixed 64 Oe ac field. The ME output values are found to be 27.5 mV/cm at fixed 3 Oe and 160 mV/cm at fixed 54 Oe ac fields. The maximum ME output obtained is 173.5 mV/cm at a fixed 64 Oe ac magnetic field (1.008 kHz frequency) and 0.7 kOe dc magnetic field. The value of ME output is higher than that reported in literature for 40% $Ni_{0.97}Co_{0.03}Mn_{0.01}Fe_{1.9}O_4$ + 60% $BaTi_{1.02}O_{3.04}$ [19].

Magnetoelectric effect in metal PZT composites is found to be higher than particulate composites. Moreover, the fabrication of composites in 2-2 laminates is found to be effective for realizing higher magnetoelectric output. The ME output in these composites is attributed to effective transfer of stress from magnetostrictive phase to piezoelectric phase due to face to face contact of two phases. The effective mechanical coupling resulted in higher charge transfer from piezoelectric phase to metal for short duration of time. Due to this metal being highly conducting phase, ME output is found to be constant in the dc bias field, whereas in the ac bias field higher charge accumulation was observed. The combination is observed to be metal/insulator/metal capacitor configuration with charge being accumulated due to bias ac field [20].

Using the above low field ME set-up (described in "Design and fabrication of ME set-up for low fields" section), ME measurements were carried out as function of ac and dc magnetic fields at room temperature. Figure 3 shows the ME output data for increasing and decreasing dc bias magnetic field for the sample Ni/PZT/Ni [1:1:1] tri-layer composite. As the dc bias field increases from 0 to +500 Oe in positive direction, +500 to decreases to −500 Oe through 0 Oe, then increased −500 to 0 Oe. A fixed 3 Oe ac magnetic field (f = 1.008 kHz) was superimposed on the dc bias field. As the dc magnetic field increases from zero, the ME output is found to increase and reaches a maximum at 60 Oe, with further increase in dc magnetic field, ME output shows a decrease. The ME output recorded was 40.52 mV/cm at 60 Oe dc bias field and 3 Oe (f = 1.008 kHz) ac magnetic field. This reveals

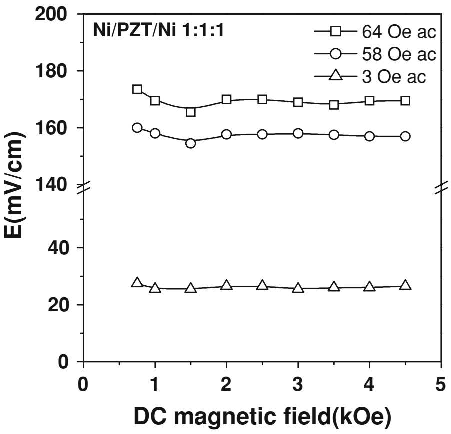

Fig. 2 Variation of ME output (E) as a function of dc magnetic field for Ni/PZT/Ni 1:1:1 laminates

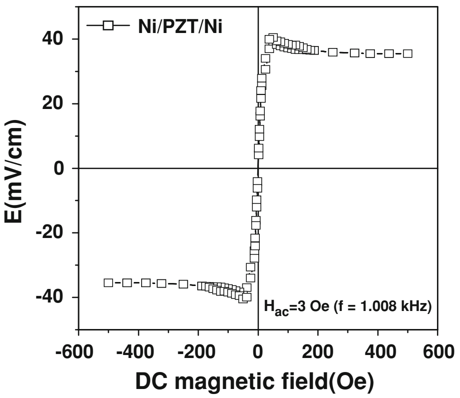

Fig. 3 Variation of ME output (E) as a function of dc magnetic field for Ni/PZT/Ni 1:1:1 laminates using low field ME set-up

the ME output dependence on the low dc and ac magnetic fields. The Ni/PZT/Ni composite showed no hysteresis, indicating low loss behaviour of ME composite in the bias magnetic field.

Latetsin et al. [16] have observed magnetoelectric output of 50 mV/cm at 600 Oe dc bias magnetic field in permendur/PZT/permendur trilayers. The value has rapidly decreased close to zero within the measured dc bias field of 1.5 kOe. The ME output is observed to vary with frequency reaching highest value at their resonance frequency. While the ME output recorded in our Ni/PZT/Ni laminate is 40.52 mV/cm at 60 Oe dc bias field (at 3 Oe ac field with frequency of 1.008 Hz) comparable to above sample and the value is constant in the measured dc bias magnetic field.

Zhang et al. [17] fabricated $Tb_{1-x}Dy_xFe_{2-y}$–Pb(Zr,Ti)O_3 (TDF–PZT) bilayers and studied the single field drive ME effect. They reported magnetoelectric voltage 330 V/cm in TDF/PZT bi layers near 5 kOe dc bias fields. The value is invariant under higher applied dc bias field (>5 kOe). The magnetoelectric output observed in TDF/PZT laminates is comparably higher than our Ni/PZT/Ni sample. This is due to the fact that the magnetostriction of TDF is around 1,600 ppm, which is almost 40 times higher than magnetostriction of Ni ($\sim$34 ppm). The TDF–PZT sample showed the maximum ME voltage value at 5 kOe dc bias field, and the value decreased with frequency reaching plateau near 10 kOe. However, our composite Ni/PZT/Ni showed a maximum ME voltage at 60 Oe dc bias magnetic filed and remains constant with varying dc magnetic field in the measured dc magnetic field range. Such behaviour is useful for exploiting the material as ac field probe under varying dc fields (<180 Oe). The material has excellent linear dependence of ac ME output with ac magnetic field under constant dc fields (60–180 Oe). This result demonstrates the utility of this composite ac magnetic field sensor in low dc bias fields.

Conclusions

Finally, it is concluded that the ME output can be realized in the laminates of Metal/FE composites, and both the participating phases need not be oxides. The study yields an ME output of 173.5 mV/cm for Ni/PZT/Ni composite at 0.7 kOe dc bias field and fixed 64 Oe ac field (1.008 kHz). Low dc magnetic field magnetoelectric set-up was fabricated and successfully utilized to study ME output of Ni/PZT/Ni system. By using the low field set-up, the ME output recorded was 40.52 mV/cm for Ni/PZT/Ni [1:1:1] composite at 60 Oe dc bias field and 3 Oe ($f = 1.008$ kHz) ac magnetic field.

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