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The influence of inducing magnetic field on the magnetoelectric effect of particulate magnetoelectric composites

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ABSTRACT

The magnetoelectric effect in a three-phase particulate magnetoelectric composite is experimentally studied with specific interest in the dependence on the inducing magnetic field up to 300 Oe. A particulate magnetoelectric composite consisting 30 vol% of PVDF, 63 vol% of PZT and 7 vol% of Terfenol-D is fabricated in lab followed by the inducing magnetic field along the diameter direction in the process of hot pressing. It is observed that the change of inducing magnetic field influences the value of the magnetoelectric coefficient. This is partly attributed to the oriented domain walls promote the increase of the magnetostrictive coefficient of Terfenol-D, and partly attributed to the contraction deformation of Terfenol-D which causes the decrease of the coupling coefficient between PZT, TD and PVDF, which together influence the change of the magnetoelectric coefficient. In addition to the inducing magnetic field, the influence of the bias magnetic field in the process of testing to the magnetoelectric coefficient is examined at different fixed resonance frequency. A maximum of magnetoelectric coefficient 82.58 mV/(cm Oe) is obtained when the inducing magnetic field H_i = 200 Oe and the bias magnetic field H_{dc} = 1000 Oe. When H_i < 200 Oe, the increase of the magnetostrictive coefficient of Terfenol-D plays a leading role which leads to increasing magnetoelectric response. In the H_i range of 200 Oe < H_i < 300 Oe, the decrease of the coupling coefficient between PZT, TD and PVDF plays a leading role and the magnetoelectric coefficient drops along the increase of the inducing magnetic field.

1. Introduction

Magnetoelectric (ME) effect is a phenomenon that generates electrical response (ME output) by external magnetic field (called ME_H effect) or magnetization by external electric field (called ME_E effect) [1–3]. Magnetoelectric materials have attracted great interest in the recent years because of their academic significance and potential applications in transducers, actuators, optoelectronics, microwave electronics and other sensors [4–6]. It is well established that the magnetoelectric effect is remarkably dependent on the magnetostrictive property, piezoelectric property and the coupling between different phases, so the magnetoelectric effect could be substantially enhanced by properly enhancing the piezoelectric and magnetostrictive properties of the composites or strengthening the magnetoelectric coupling [7].

To obtain high piezoelectric property, ceramics such as $BaTiO_3$ and lead–zirconate–titanate (PZT) have been always chosen as the piezoelectric phases in $CoFe_2O_4/BaTiO_3$, $CoFe_2O_4$/lead–zirconate–titanate (PZT), Tb–Dy–Fe alloys (Terfenol-D)/PZT composites [8–11]. After the polymer was introduced into magnetoelectric composites to overcome brittleness and high eddy current loss, PVDF and [P(VDF-TrFE)] [12–14] were applied for their piezoelectricity. Moreover, the unanimous polarization of piezoelectric ceramics and polymer have been realized by the high electric field, which leads to optimized piezoelectric property. The magnetoelectric coupling of different phases is of great importance in stress or tension transmission which contributes to the enhancement of magnetoelectric effect. To obtain a high coupling, Nan and co-workers have taken polymer PVDF as the binder in a kind of three-phase composites [15]. Viehland and co-workers have prepared a kind of laminate composite that different phases are bonded together by silver epoxy [16]. Martins and his groups have prepared ferrite/PVDF nanocomposites that ferrite and PVDF are coupling by anionic surfactants (SDS) [17] etc. To obtain high magnetostrictive property, most researchers have always chosen $CoFe_2O_4$, Terfenol-D and Metglas as the magnetostrictive phase in $CoFe_2O_4/BaTiO_3$, (Terfenol-D)/PZT, Metglas/PVDF [18–21] etc., however, few researchers have focused on the influence of the magnetic field on the magnetoelectric effect which may influence the magnetostrictive property or magnetoelectric coupling. Compared with electric field, magnetic field can affect the alignment

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and configuration of material without contact. Moreover, magnetic field might have significant effect on the property of powder especially ferromagnetic particles under special circumstances such as high temperature and pressure situation.

In the present work, we have to study the influence of a stable inducing magnetic field on magnetostrictive property of magnetic phase, the magnetoelectric coupling of different phases and the magnetoelectric effect of particulate magnetoelectric composites in the process of hot pressing molding. Our composite structure consists of Tb–Dy–Fe alloy (Terfenol-D) used as magnetostrictive phase, lead zirconate titanate (PZT) used as piezoelectric phase and poly vinylidene fluoride (PVDF) used as binder and stress or tension transfer medium. This kind of composite is chosen in the work for its easy fabrication, good flexibility and precise control of the process parameters.

2. Experimental procedure

Terfenol-D powder with a mean particle size of 20 μm was obtained by crushing bulk $Tb_{0.28}Dy_{0.72}Fe_{1.95}$ single crystal (Gansu Tianxing Rare Earth Functional Materials Co., Ltd.) in an argon atmosphere. PZT powder with a mean particle size of 5 μm was prepared via the conventional solid state reaction method. N,N-dimethylformamide (DMF, pure grade) was supplied by Fluka. In order to obtain a good dispersion of the Terfenol-D and PZT particles within the polymeric matrix, the following procedure was implemented. Firstly, 10 g PVDF (Sichuan Chenguang chemical research institute) was dissolved to the corresponding amount of DMF (The ratio of mass of PVDF to volume of DMF is 1 g: 8 ml), and mixed in a Teflon mechanical stirrer for 1 h. Secondly, the corresponding to the volume of Terfenol-D and PZT were added to the PVDF mixed solution, and mixed in a Teflon mechanical stirrer for 3 h. The flexible thick films were obtained by tape-casting method on a clean glass substrate. The thickness of the thick film was about 30 μm, after solvent was evaporated at 100 °C for 2 h. The thick films were cut into pieces and molded by pressing at 190 °C under a pressure of 10 MPa. As shown in Fig. 1, the inducing magnetic field H_i has been applied continuously to the sample in the whole process of hot pressing. Disk-shaped samples of diameter 15 mm were cut from the molded sheet (1 mm in thickness) and silver-paint electrodes were applied to the samples. The samples were poled in silicone oil under a poling field of 2–5 kV/mm and at a poling temperature of 100 °C. The thickness direction is the direction of the polarization of the sample (denoted as X_3 axis of the samples).

The dielectric properties of the samples were measured using a HP4192A impedance analyzer. The piezoelectric constant d_{33} was measured by a standard piezo d_{33} meter. Magnetostriction measures were performed by the standard strain-gauge technique. In order to measure the magnetoelectric coefficient, an electromagnet was used to provide the bias field up to 3000 Oe along the diameter direction (denoted as X_1 axis of the samples), which was also the direction of the inducing magnetic field. The coefficient was measured directly as response of the sample to an AC magnetic input signal at 100 Hz and about 2 Oe amplitude superimposed on the DC bias field, both parallel to the X_1 axis. A signal generator (DG4102, Beijing, China) was used to drive the Helmholtz coils and to generate the AC magnetic field. The voltage generated from the particulate composites was measured under an open circuit condition by using a differential amplifier (DHF-2, Beijing, China). The output signal from the amplifier was measured with a digital oscilloscope (MSO1104Z-S, Beijing, China). All measurements were performed at room temperature. The magnetoelectric coefficient of the magnetoelectric composites can be given by Eq. (1).

$$\alpha_{ME} = E/H = U/(d \cdot H) \quad (1)$$

3. Results and discussion

3.1. The influence of each phase's content in PZT/PVDF/Terfenol-D composites

The piezoelectric properties of PVDF/PZT composites in which the volume fraction of PVDF was range from 0 to 0.5 were measured. Fig. 2 shows the piezoelectric property d_{33} decreases with the increase of PVDF, high concentration of PVDF led to weak piezoelectric properties; on the other hand, low volume fraction of PVDF made it difficult to fabricate the composite. When the volume fraction of PVDF was 0.3, the piezoelectric property and flexibility achieve a state of equilibrium, so the volume fraction of PVDF was fixed as 0.3 in the following experiments.

In the three-phase composites PVDF/PZT/Terfenol-D, the volume fraction of PVDF was fixed as 0.3. The particulate composites of 0.3 PVDF/$f_{\text{Terfenol-D}}$Terfenol-D/(0.7-$f_{\text{Terfenol-D}}$)PZT were prepared where $f_{\text{Terfenol-D}}$ denotes the volume fractions of Terfenol-D. Fig. 3 showed that PZT and Terfenol-D particles well distributed in the PVDF matrix. Fig. 4 showed the piezoelectric properties and the maximum magnetoelectric coefficients with the bias magnetic field H_{dc} = 1000 Oe measured for the three-phase composites. When $f_{\text{Terfenol-D}} < 0.07$, the magnetoelectric coefficients increased with the increment of $f_{\text{Terfenol-D}}$, however, when $f_{\text{Terfenol-D}} > 0.07$, the piezoelectric properties changed dramatically for the low resistance Terfenol-D particle percolation path can make the samples difficult to polarize, and the magnetoelectric coefficients dropped dramatically because of the weak piezoelectric properties.

So we chose $f_{\text{Terfenol-D}} = 0.07$ as the volume ratio of Terfenol-D in the following experiments.

3.2. The influence of the inducing magnetic field H_i to the resonance frequency

The magnetoelectric coefficient is experimentally obtained for the 0.3PVDF/0.63PZT/0.07Terfenol-D particulate magnetoelectric composites with bias magnetic field in the X_1 direction. Fig. 5

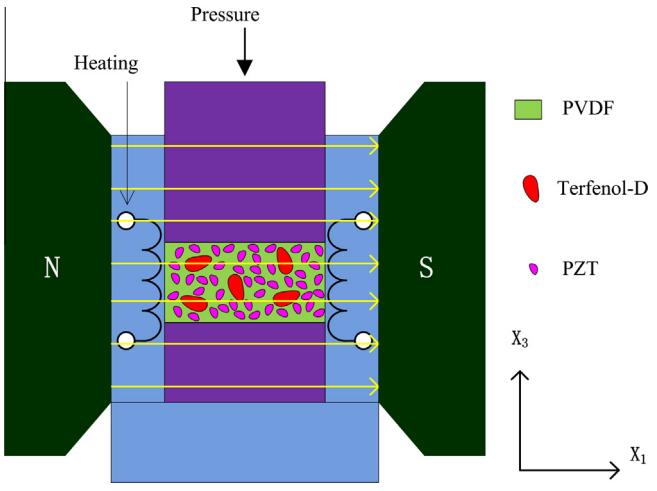

Fig. 1.

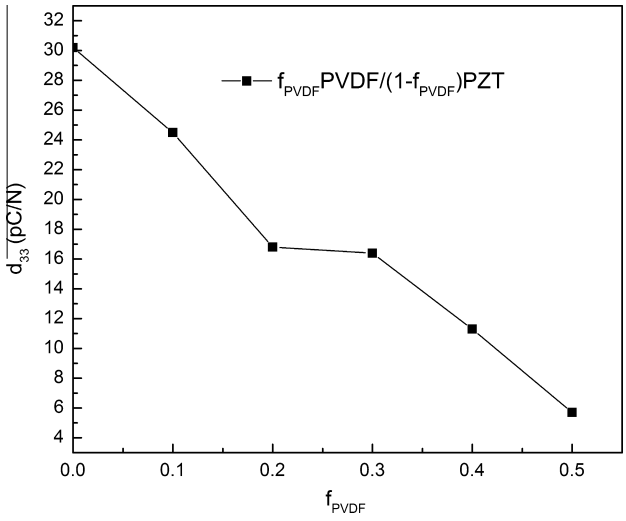

Fig. 2.