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Electrospun barium titanate/cobalt ferrite composite fibers with improved magnetoelectric performance

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In this study, we use a versatile sol-gel based electrospinning technique to fabricate nanostructured barium titanate (BaTiO$_3$)/cobalt ferrite (CoFe$_2$O$_4$) composite fibers and analyze their magnetoelectric response. Scanning and transmission electron microscopy images indicate that the obtained fibers are composed of fine grains which are self-assembled and arranged. X-ray diffraction (XRD) study of the composite fibers revealed the presence of perovskite and spinel structures corresponding to BaTiO$_3$ and CoFe$_2$O$_4$ phases, respectively. The magnetic hysteresis loops of the resultant fibers showed that the fibers were ferromagnetic with magnetic coercivity of 650 Oe and saturation magnetization of 31 emu/g. Moreover, the magnetoelectric coefficient developed on the surface of the fibers was measured as a function of the applied external DC magnetic field. A maximum magnetoelectric coefficient of 13.3 mV.cm$^{-1}$Oe$^{-1}$ was determined for the composite fibers, which is much higher than the commonly reported values for bulk or thin film counterparts. The large magnetoelectric coefficient of the composite fibers was attributed to the nano-sized grains and their arrangement within the fibrous geometry. The intimate contact and the large interfacial area presented by the nanostructures ensured that the composites displayed strong magnetoelectric behavior. Such composite fibers show tremendous potential for magnetic field sensor applications and for magnetoelectric devices.

**Introduction**

Multiferroic materials with coexistence of two or more ferroic orders such as ferroelectricity, ferromagnetism and ferroelasticity have attracted a great deal of interest due to their potential applications as multifunctional devices. In some multiferroic materials, the interaction between the magnetostrictive and ferroelectric phases gives rise to a new effect called the “magneto-electric (ME)” effect. The ME effect makes it possible for the electrical polarization/magnetization within the material to be controlled with the application of external magnetic/electric field. This desired mechanism enables a variety of potential applications for ME materials including sensors, transducers, memory devices which can be electrically written and magnetically read, magnetically controlled piezoelectric devices, etc.

The numbers of monophasic multiferroic materials that are capable of displaying the ME effect are limited because it is difficult for the electric and magnetic dipoles to coexist at ambient temperatures within the asymmetrical structures. Furthermore, the ME effect in monophasic materials is usually very small. Hence, many efforts have been directed towards fabrication of two-phase hetero-structures. ME composites consisting of two phases offer greater flexibility, multifunctionality, and higher coupling compared to naturally occurring monophasic materials. The ME effect in such composites depends on the magnetic-strain-electrical coupling between the magnetostrictive and ferroelectric phases. When a magnetic field is applied, the magnetostrictive phase within the composite induces a strain in terms of a change in dimensions, which in turn transfers the stress to the ferroelectric phase, resulting in an electric polarization. This strong coupling enables composites capable of large ME coefficients. However, obtaining good interfacial bonding between the phases is a prerequisite for fabricating composites which are capable of displaying significant ME effects. Composites fabricated using conventional techniques, such as sintering, microwave sintering and hot pressing are found to exhibit low ME coefficient due to the presence of micro-cracks, impurity phases, non-ideal interfaces between the phases, defects, and high leakage currents. To address this issue, recent studies have focused on fabricating nanostructured composites. It has been demonstrated that the nanometer length scale of the constituent phases within the composites ensures strong interfacial coupling and intimate contact between the phases.

Herein, we report the fabrication of nanostructured composite fibers using a sol-gel enabled electrospinning technique, hitherto unexplored, and demonstrate that these fibers display well-defined ME behavior. Indeed, very few studies have explored the magnetoelectric behavior of electrospun fibers. These studies used piezoresponse force microscopy (PFM) to investigate the local magnetoelectric coupling of the fibers.
imposed on the fibers to confirm the presence of ME coupling. Here, we obtain BaTiO$_3$/CoFe$_2$O$_4$ composite fibers using electrospinning enabled techniques and investigate the ME coefficient of the fibers as opposed to using PFM to probe the local ME coupling and estimating their ME coefficient. The ME coefficient values for bulk composite typically vary between 0.19 to 4 mV cm$^{-1}$ Oe$^{-1}$. We demonstrate that electrospinning is not only useful to fabricate BaTiO$_3$/CoFe$_2$O$_4$ composite fibers but it can also help composites that display sizeable ME coefficients. BaTiO$_3$ and CoFe$_2$O$_4$ are chosen in this study as they are individually known to display good ferroelectric and ferromagnetic behaviors, respectively. In the composite fiber form, they combine the electric and magnetic polarizations to exhibit a prominent ME effect. These one-dimensional ME fibers are suitable for many applications, such as microelectronic devices, electromagnetic devices and nano-systems among others.

Experimental

BaTiO$_3$/CoFe$_2$O$_4$ composite fibers were fabricated using the sol-gel enabled electrospinning technique. For this purpose, BaTiO$_3$ and CoFe$_2$O$_4$ precursor sol-gel solutions were separately processed before preparing the electrospinning solution. First, BaTiO$_3$ precursor solution referred to as solution A was prepared by dissolving 2.55 g of barium acetate in 6 ml of acetic acid. 2.95 ml of titanium isopropoxide was added to this solution under constant stirring condition. Following this, CoFe$_2$O$_4$ precursor solution referred to as solution B was prepared by dissolving 2.9 g of Co(NO$_3$)$_2$.6H$_2$O and 8.08 g of Fe(NO$_3$)$_3$.9H$_2$O in 10 ml of dimethylformamide (DMF) solution. Solution A and B were added in a ratio in order to obtain a molar ratio of 1:1 between CoFe$_2$O$_4$ and BaTiO$_3$.

Electrospinning

The solution for electrospinning was prepared by dissolving 3 g of polyvinyl pyrrolidone (PVP) with molecular weight 360,000 in 11 g of DMF/ethanol solvent mixture (1:1 wt/wt). Solution A was added to the PVP solution drop-wise and under constant stirring condition; then, solution B was added drop by drop. The mixture solution was stirred for 24-36 h until a homogenous solution was obtained.

A syringe with a 23-gauge metal needle was used to electrospin the mixture solution. Electrospinning was conducted at 21 kV and the flow rate of the solution was 0.07 mm/min. 15 cm spacing was used between the tip of the needle and the collector surface.

The obtained fibers were initially dried in an oven at 100 °C for 24 h and subsequently transferred to a furnace for thermal annealing. The fibers were heated from ambient temperature to 750 °C. The heating rate was 5 °C/min and the dwelling time at 750 °C was 1 h.

Structural characterization

Microstructures

The morphology of the fibers was analysed using a field emission scanning electron microscope (FESEM, Zeiss ULTRA plus). The precursor fibers and the BaTiO$_3$/CoFe$_2$O$_4$ fibers obtained after thermal annealing were coated with gold before they were examined using a SEM at an accelerating voltage of 5 kV. Transmission electron microscopy (TEM, Philips CM120 Biofilter) was used to examine the BaTiO$_3$/CoFe$_2$O$_4$ fibers.

X-ray diffraction (XRD)

The crystal structures of the fibers were determined from the XRD patterns of the BaTiO$_3$/CoFe$_2$O$_4$ fibers. XRD on the BaTiO$_3$/CoFe$_2$O$_4$ samples were conducted in reflection mode at ~25 °C using an X-ray diffractometer (XRD Shimadzu S6000) with CuKα radiation (λ=1.54 Å). The 20 scan was varied between 15° and 70° and the scan speed was set at 1°/min with 0.02° step size.

Ferromagnetic behavior

Measurements of the room temperature ferromagnetism for BaTiO$_3$/CoFe$_2$O$_4$ fibers were recorded using a commercial Quantum Design magnetic property measurement system (MPMS) and physical property measurement system (PPMS). A vibrating sample method was used to obtain the magnetic hysteresis curves. The magnetic field was ramped at ambient temperature (300 K) from 10,000 Gauss to -18,000 Gauss and back to 18,000 Gauss.

Magnetoelectric effect

To measure the ME response, the fibers were first ground and broken into shorter fragments which were then pressed together under a pressure of 130 MPa and sintered at 1100 °C for 1 h to obtain disk shaped specimens. 5 °C/min was used as the heating rate. The ambient temperature ME effect on the sintered disk specimens was determined using the dynamic lock-in technique. The measurement was performed at a constant frequency of 1 kHz and a constant bias AC magnetic field (5 Oe). The ME coefficient of the samples was measured as a function of the DC magnetic field (H$_{DC}$) and was obtained by dividing the measured output voltage by the magnitude of applied H$_{AC}$ and the thickness of the sample disk.

Results and discussion

Typically, introduction of indirect coupling through strain between the ferroelectric and ferromagnetic phases within the composite ensures that the composite displays a ME effect. Good coupling and optimized ME response in composites can be obtained by controlling the size of the constituent phases and the quality of the interfacial bonding between them. Thus, composite samples with nanostructured constituent phases can display large ME properties since the nanometer length scale of the constituents offers large interfacial area. In this study, we fabricate barium titanate (BaTiO$_3$)/cobalt ferrite (CoFe$_2$O$_4$) composite fibers using a sol-gel based electrospinning technique. Our results demonstrate that these fibers composed of nanostructured BaTiO$_3$ and CoFe$_2$O$_4$ grains display well-defined ferromagnetism and ME coefficient.

The surface morphology of as-spun fibers and fibers obtained after thermal annealing is evaluated using SEM. Figures 1A and 1B show such images of as-spun fibers and BaTiO$_3$/CoFe$_2$O$_4$ fibers, respectively. The as-spun precursor fibers in Figure 1A show an average fiber diameter of 300 nm with a smooth fibrous
morphology. After thermal annealing, brownish black fibers, tens of micrometers in length, are formed. It is clear from Figure 1B that the resultant BaTiO$_3$/CoFe$_2$O$_4$ fibers retained their fibrous morphology. However, the diameter of BaTiO$_3$/CoFe$_2$O$_4$ fibers is reduced to 140±30 nm, which is attributed to the decomposition of the polymer and crystallization of BaTiO$_3$/CoFe$_2$O$_4$.

Figure 2 shows typical TEM images of a BaTiO$_3$/CoFe$_2$O$_4$ composite fiber. It is evident that the fiber is composed of fine nanoparticles that are linked and tightly bound. Figure 2B shows that the linked particles display interference of two crystallographic orientations, which originate from the crystals of BaTiO$_3$ and CoFe$_2$O$_4$. A magnified image using Digital Micrograph software identified the crystallographic phases by measuring the inter-planar spacing, d. BaTiO$_3$ is identified by the (100) plane, while CoFe$_2$O$_4$ the (220) plane. Figure 2B also demonstrates that the BaTiO$_3$ and CoFe$_2$O$_4$ particles are in close proximity with each other. These results confirm the formation of crystalline BaTiO$_3$/CoFe$_2$O$_4$ composite fibers.

X-ray diffraction patterns of the samples are also obtained to confirm the crystalline phases corresponding to BaTiO$_3$ and CoFe$_2$O$_4$. The XRD patterns of the as-spun precursor fibers (not shown in Figure 3) do not show any peaks, which indicate that the as-spun fibers are amorphous. However, XRD patterns of the thermally annealed sample (Figure 3) shows clear distinct peaks, confirming that the sample is polycrystalline. The diffraction pattern peaks are indexed to the reflections of BaTiO$_3$ and CoFe$_2$O$_4$ phases. This illustrates that the BaTiO$_3$ and CoFe$_2$O$_4$ phase evolution occurs from the amorphous solid phase and during the thermal annealing process. The phases are distinct and appear well segregated. All the reflections can be indexed according to the structures of BaTiO$_3$ and CoFe$_2$O$_4$ phases respectively (JCPDS card number 81-2196 and JCPDS card number 22-1086). The presence of a few unidentified peaks suggests some form of chemical reactions between BaTiO$_3$ and CoFe$_2$O$_4$ phases during sintering. The peak seen at 37° corresponds to the Co$_3$O$_4$ impurity phase (JCPDS card number 42-1467).

Next, we investigate the multiferroic properties of the fabricated composite fibers by measuring their ferromagnetic and direct ME coupling behavior. The magnetic hysteresis loop of the composite fiber measured along the in-plane direction and at room temperature is given in Figure 4 and is typical of magnetic materials. This indicates the existence of an ordered magnetic structure in the fibrous composite. The saturation magnetization of the BaTiO$_3$/CoFe$_2$O$_4$ composite fiber is determined from the intercept to the y-axis (magnetization) as 1/H approaches zero. Hence, the saturation magnetization ($M_s$) of
between ferromagnetic and anti-ferromagnetic materials result in the displacement of the hysteresis loops. Our samples are composites consisting of a ferrite phase and a ferroelastic phase. These phases are randomly dispersed within the fiber geometry, causing lattice distortion of the ferrite phase. Hence, the stress generated by this lattice distortion and magnetoelastic coupling between the phases may be responsible for the displaced hysteresis loops. The presence of the piezoelectric phase within the composite should also play a role in influencing the hysteresis behavior. This is also supported by the results of Rizwan and co-workers. They suggest that the coupling between the ferromagnetic and piezoelectric domains and the hindrance to the magnetic ordering of the ferrite phases provided by the ferroelastic phase lead to the asymmetrical hysteresis loops. Study of the exact mechanism responsible for the displaced M-H hysteresis loop is beyond the scope of this work. The coercive field for asymmetric loops can be defined by: \( H_c = \frac{(H_{c2} - H_{c1})}{2} \), where \( H_{c1} \) and \( H_{c2} \) are the right and left coercive fields of the hysteresis loop. Thus, the coercive field is determined to be ~670 Oe. This magnetic coercive field depends on several factors including magneto-crystalline anisotropy, grain size, interface, defects, doping and nature of the surface. The low value of the coercive field recorded at room temperature indicates that the BaTiO₃/CoFe₂O₄ composite fibers are magnetically soft and show tremendous potential for device applications. However, the coercive field determined for BaTiO₃/CoFe₂O₄ composite fibers is slightly higher than the theoretical coercive field 430 Oe of CoFe₂O₄. We attribute this to the presence of the BaTiO₃ phase, which is a non-magnetic phase and makes the magnetization difficult.

The saturation magnetization of these composite fibers obtained using the electrospinning technique is found to be higher than the saturation magnetization of some of the ferrite/ferroelectric composite systems reported in. The higher Mₛ of our composite fibers can be attributed to the unique processing method. Similar results are obtained by Arias et al. They show that electrospinning leads to elongated grains of the ferrite fibers possessing enhanced magnetic properties. These results are well supported by theory, which predicts that fibrous systems having grains arranged and aligned in a linear chain configuration will have their magnetic dipoles also aligned along the fiber axis. The interaction between the dipoles of neighboring grains and the alignment of dipoles contribute towards the enhancement of saturation magnetization. However, the \( M_s \) value for the composite fibers is lower than the theoretical value of 71 emu/g for pure CoFe₂O₄. This is caused by the presence of the non-magnetic BaTiO₃ phase within the composite.

Figure 5 shows the dependence of the ME coefficient of the composite fibers on the DC magnetic field. The ME coefficient recorded for the composite fibers is a measure of changes in the electric voltage in response to the applied external magnetic field. During the measurements, a small AC magnetic field (\( H_{AC} \)) of 5 Oe is used in conjunction with the DC magnetic field (\( H_{DC} \)). It is evident from Figure 5 that the BaTiO₃/CoFe₂O₄ composite fibers have a small initial ME coefficient value near zero magnetic bias. At zero magnetic field, the ME coefficient of the composite fibers is ~1.7 mV.cm⁻¹.Oe⁻¹. As the magnetic field is increased, the ME coefficient of the composite fibers shows an approximate linear increase with magnetic field and attains a maximum value of 13.3 mV.cm⁻¹.Oe⁻¹ at DC magnetic field of ~1810 Oe. At magnetic field greater than 1810 Oe, the ME coefficient value saturates and then decreases with magnetic field. The value (13.3 mV.cm⁻¹.Oe⁻¹) at which ME saturates is taken as the ME coefficient of the composite fibers.

Table 1 summarizes the results of some current CoFe₂O₄/BaTiO₃ ME composites. Evidently, the ME coefficient we obtained is much larger than those commonly reported on ME coefficient of BaTiO₃/CoFe₂O₄ composites prepared from core-shell powder or powder and comparable to those containing nano-lamellar bicrystals. This indicates strong coupling between the ferroelectric (BaTiO₃) and ferromagnetic
(CoFe$_2$O$_4$) phases in our fabricated composite fibers. Differences in ME coefficient values reported in the literature may be due to differences in sample characteristics such as constituents, microstructure, size, etc.$^{10,41}$. For example, coupling between the phases can be improved by increasing the sintering temperature. However, high sintering temperature can lead to the formation of Fe$^{2+}$ ions in the ferrite phase. These Fe$^{2+}$ ions can reduce the electrical resistivity of the composite, which is important to achieve a high ME signal.$^{16}$ Nie et al.$^{41}$ reported a high ME coefficient value of $\sim 17$ mV.cm$^{-1}$Oe$^{-1}$ for their composite particles. They sintered the samples at 1200 °C for 2 h and were able to control the crystallite size of their composite between 35 and 80 nm. Furthermore, their samples were poled before the ME measurements were made. However, low ME values ($0.3$ to 2 mV.cm$^{-1}$Oe$^{-1}$) were also reported in other studies.$^{10,11,38}$ for similar composites. This could be because of the high sintering temperature and the duration for sintering used for sample preparation. For example, Corral-Flores et al.$^{38}$ sintered the samples at 1200 °C for 12 h. In addition, the sizes of BaTiO$_3$ and CoFe$_2$O$_4$ in the composite were much larger at $\sim 2$ μm. These factors could explain the differences in ME coefficient values reported for composite particles in References 41 and 38.

Studies which have reported even higher ME coefficient values fabricated composites by dispersing CoFe$_2$O$_4$ nanorods within the BaTiO$_3$ matrix phase. The dispersion of single crystal nanorods within the BaTiO$_3$ matrix played a role in enhancing the interfacial area between the ferroelectric and ferromagnetic phases and also ensured good ME coupling between these two phases.$^{12,42}$ For example, Bai et al.$^{43}$ reported a ME coefficient value of $1.2 \times 10^2$ mV.cm$^{-1}$Oe$^{-1}$. Such a high ME coefficient value could be attributed to the large interfacial area obtained by dispersing CoFe$_2$O$_4$ nanorods within the BaTiO$_3$ phase. Also, the ME coefficient values were obtained using piezoresponse force microscopy (PFM), which only measured the local ME response. BaTiO$_3$ phase was electrically poled prior to ME measurements to increase its ferroelectric response. It is well-known that when the PFM tip contact the sample surface for measurements, it exerts a compressive stress of $\sim 100$ MPa.$^{44}$ This stress is large enough to induce strains in the local piezo/ferro electric domains, which has an effect on the measured ME coefficient. Similarly, Deng et al.$^{45}$ used hydrothermal reaction and polymer assisted deposition to fabricate single crystal CoFe$_2$O$_4$ nanorods embedded in BaTiO$_3$ matrix and obtained a ME coefficient of 51 mV.cm$^{-1}$Oe$^{-1}$. This quite high ME coefficient value obtained is attributed to the large interfacial area induced by the nano-sized CoFe$_2$O$_4$ and BaTiO$_3$ phases and ‘perfect interface’ achieved by using polymer assisted deposition. It should be noted that multi-layered BaTiO$_3$/CoFe$_2$O$_4$ composites$^{45}$ give the lowest ME coefficient value.

It is well-known that the ME effect in composites is a product tensor property and is typically affected by (i) the geometry and morphology of the phases, (ii) interfacial bonding between the phases, (iii) individual properties of each constituent phase, and (iv) thickness and number of magnetostrictive and ferroelectric phases.$^{23,33}$ In our work, we believe the increased ME coefficient is a direct result of the unique electrospinning process. Electrospinning followed by thermal annealing of precursor fibers leads to the formation of composite fibers composed of BaTiO$_3$ and CoFe$_2$O$_4$ grains. The grain sizes of individual phases are in the range of tens of nanometers. Nanoscale sizes of these grains can induce large interfacial area, which will favour elastic interactions between the ferroelectric and ferromagnetic phases. It is also known that large interfacial area and good phase connectivity play a role in contributing to the strong ME effect. Moreover, it is clear from the TEM images in Figure 2 that the obtained fibers have BaTiO$_3$ and CoFe$_2$O$_4$ in close proximity. This ensures that the phases are tightly bound and the strains can be readily transferred to the neighboring grains. Hence, the phase-connectivity of these nanostructures also contributes towards the ME effect of the composite.

Further increase in magnitude of the magnetic field beyond the critical value of 1810 Oe leads to decreasing ME coefficient values. Ferromagnetic materials can change their shape during the process of magnetization and this property is called magnetostriction. As the magnetic field is increased, the magnetostriction increases gradually and attains a maximum value. During this stage, the strain produced in the ferromagnetic material is transferred to the ferroelectric BaTiO$_3$ phase due to the coupling between the ferromagnetic and ferroelectric phases. The ferroelectric phase produces a voltage in response to the strain. Beyond $\sim 1810$ Oe, the magnetostriction and the strain produced in the composite would produce a constant electric field in the ferroelectric phase. Beyond saturation magnetization, the magnetostriction and the strain thus produced in the CoFe$_2$O$_4$ phase generates a constant electric field in the piezoelectric BaTiO$_3$ phase.$^{39}$ This suggests that change of electric field ($dE$) per applied magnetic field change ($dH$), $dE/dH$, which is a measure of magnetoelastic coefficient decreases as the magnetic field is increased beyond the critical value of $\sim 1810$ Oe. Thus, the magnetostrictive coefficient reaches saturation at $\sim 1810$ Oe. This explains why the ME coefficient increases initially and then begins to decrease when the applied magnetic field is increased beyond a critical value. This trend in the ME coefficient versus magnetic field can be attributed to the spinel ferrite phase of CoFe$_2$O$_4$.$^{10,11,39}$

**Fig. 5** Dependence of the magnetoelectric (ME) coefficient of BaTiO$_3$/CoFe$_2$O$_4$ composite fibers on the DC magnetic field. An AC magnetic field of 5 Oe was applied during the measurements.
Table 1 ME coefficient and saturation magnetization values of CoFe$_2$O$_4$/BaTiO$_3$ composites

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<th>No</th>
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<th>ME coefficient (mV.cm$^{-1}$.Oe$^{-1}$)</th>
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Conclusion

In summary, our results demonstrate that the BaTiO$_3$/CoFe$_2$O$_4$ fibers obtained by electrospinning possess excellent ferromagnetic and ME properties. The ME coefficient of these one-dimensional structures are found to be much higher than their bulk or thin film counterparts. The large value of the ME coefficient is attributed to the electrospinning technique which produces fibers with diameters in the range of 100-200 nm. The BaTiO$_3$ and CoFe$_2$O$_4$ grains within the fibers display sizes of few tens of nanometers. Consequently, they provide large interfacial areas and excellent interphase connectivity so that the composite fibers have large ME coefficients. These one-dimensional electrospun composite fibers show tremendous potential as novel ME materials for nanoscale devices and applications.

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Notes and references