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Electrical, magnetic, and direct and converse magnetoelectric properties of $(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3-(x)CoFe_2O_4$ (PZT–CFO) magnetoelectric composites

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ABSTRACT

In this work, hydrothermal synthesis and ceramic sintering process were applied to fabricate $(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3-(x)CoFe_2O_4$ (PZT–CFO) (x=0.20, 0.35, and 0.50) magnetoelectric (ME) composites. The X-ray diffraction (XRD) studies revealed that no chemical reaction occurred between individual PZT and CFO phases and confirmed the manifestation of PZT and CFO phases within the ME composite. Scanning electron microscopy (SEM) was used to investigate the microstructure and connectivity scheme in the ME composites. The dielectric constant (ε) and loss tangent ($\tan\delta$) were determined as functions of frequency and temperature for all the composites prepared. The polarization–electric field (P–E) and magnetization–magnetic field (M–H) hysteresis loops obtained indicate that both ferroelectric and ferromagnetic properties coexist in the ME composites prepared. The direct (DME) and converse magnetoelectric effects (CME) were measured only for the PZT–CFO20 (20 mol% $CoFe_2O_4$) composite sintered at 1100 °C for 4 h. The maximum values of ME voltage coefficient $\alpha_{ME}(dE/dH)$ and ME susceptibility coefficient α_{me} (dH/dE) were 226 mV cm^{-1} Oe^{-1} at frequency of 75.1 kHz and 1.15×10^{-8} s/m at frequency of 66.8 kHz, respectively.

1. Introduction

With the rapid development of modern technology, it is necessary to develop more advanced materials to satisfy today's high requirements (e.g. micromation, intelligence, multifunction, integration, high performance, etc.) in exploiting novel functional elements and devices. One of the most interesting categories of advanced materials is multiferroics, in which there are two or more ferroic orders, such as ferromagnetism, ferroelectricity or ferroelasticity, coexist and/or coupled with each other [1]. Important potential applications of these multiferroics include transducers, sensors, actuators, filters, non-volatile memories, etc. [2–4]. A majority of single-phase multiferroics, including Cr_2O_3 [5], $Ni_3B_7O_{13}I$ [6], YIG [7], $Pb(Fe_{0.5}Nb_{0.5})O_3$ [8], $BiFeO_3$ [9], $BiMnO_3$ [10], rare earth manganates [11–13], etc., have a weak magnetoelectric (ME) coefficient at or above room temperature, making them inadequate for practical applications. Interestingly, the ME composites combined with magnetostrictive ($CoFe_2O_4$, $NiFe_2O_4$, $CuFe_2O_4$, Tefernol-D alloys, etc.) and piezoelectric ($Pb(Zr_{0.52}Ti_{0.48})O_3$, $BaTiO_3$, PMN-PT, etc.) phases can have a desirable room temperature ME effect originated from the elastic interaction among the subsystems via a stress mediation [14–16]. Up to date, a great deal of efforts has been done in researching on ME composites with various connectivities, such as particulate composites [3,17], laminated composites [18–20], thin films [21–23], etc. Compared with thin films and laminated composites, particulate composites have advantages of low cost, simple production technology, good ME effect for favorable microstructures and easy control of electrical and magnetic properties and ME coefficient with a proper choice for ferroelectric and ferromagnetic phases and their proportion [24–26].

The hydrothermal synthesis has been widely used to synthesize nanopowders with good crystallinity, narrow particle size

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distribution, and controlled stoichiometry. Therefore, the hydrothermal synthesis of high quality ferroelectric and ferromagnetic nanopowders as the components for the fabrication of ME composites will be beneficial for obtaining high ME coupling effect. A substantial number of research have been focused on the fabrication of $Pb(Zr_{0.52}Ti_{0.48})O_3$–$CoFe_2O_4$ (PZT–CFO) composites [27–30], in which PZT with morphotropic phase boundary (MPB)

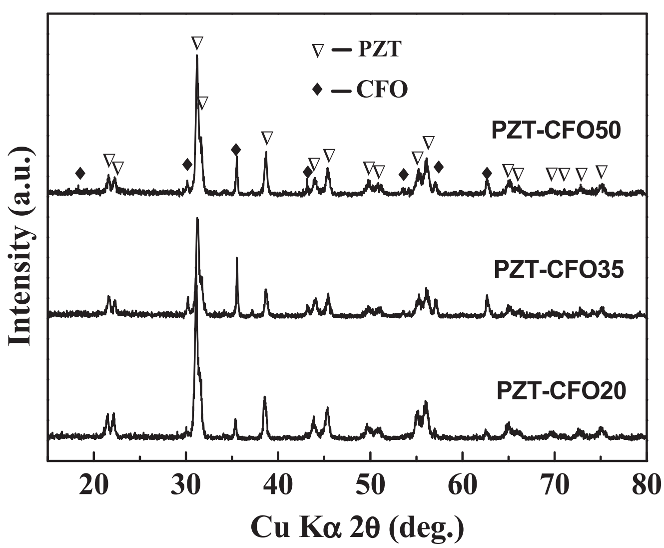

Fig. 1. XRD patterns of PZT–CFO20, PZT–CFO35, and PZT–CFO50 composites sintered at 1100 °C for 4 h.

structure was chosen as the ferroelectric phase for its excellent ferroelectric and piezoelectric properties, and CFO was chosen as the ferromagnetic phase for its large magnetocrystalline anisotropy, high coercivity, moderate saturation magnetization as well as remarkable chemical stability and mechanical hardness [31]. It is interesting to find out that the hydrothermal synthesis of PZT–CFO composite has not been reported previously.

In this work, we therefore report on the fabrication of $(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3$–$(x)CoFe_2O_4$ (PZT–CFO) (x=0.20, 0.35, and 0.50) magnetoelectric (ME) composites by hydrothermal method and ceramic sintering process. The microstructure, dielectric, ferroelectric, ferromagnetic and ME coupling properties of PZT–CFO composites were investigated.

2. Experimental

2.1. Preparation

The $(1-x)Pb(Zr_{0.52}Ti_{0.48})O_3$–$(x)CoFe_2O_4$ (PZT–CFO) (x=0.20, 0.35, and 0.50) magnetoelectric (ME) composites were fabricated by hydrothermal synthesis and ceramic sintering process. The experimental procedure for the fabrication of PZT–CFO composites includes three steps. (1) For the synthesis of $Pb(Zr_{0.52}Ti_{0.48})O_3$, $Pb(NO_3)_2$, $ZrOCl_2$, and amorphous TiO_2 were used as starting materials. First, stoichiometric amounts of starting materials were dissolved and mixed in deionized water under magnetic stirring and then KOH was introduced into the solution to adjust the pH to 3 M $[OH^-]$ under vigorous stirring. After that, the precursor suspension was transferred into a Teflon-lined stainless steel

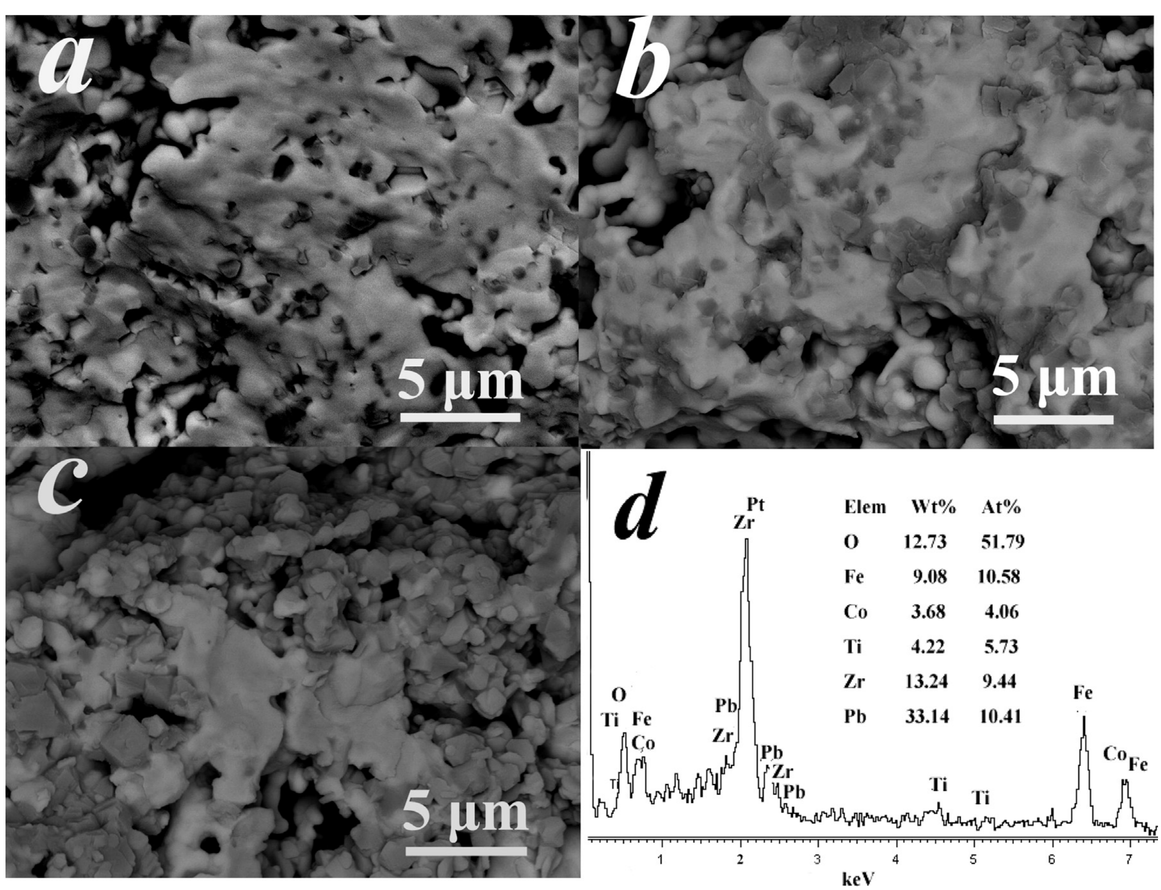

Fig. 2. Back-scattered SEM images of the fractured surface of PZT–CFO20 (a), PZT–CFO35 (b), and PZT–CFO50 (c) composites sintered at 1100 °C for 4 h and EDS spectrum of PZT–CFO35 composite (d).