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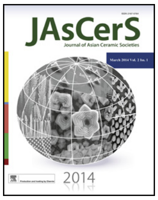

Full Length Article

Strong and anisotropic magnetoelectricity in composites of magnetostrictive Ni and solid-state grown lead-free piezoelectric BZT–BCT single crystals

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ABSTRACT

Aimed at developing lead-free magnetoelectric (ME) composites with performances as good as lead (Pb)-based ones, this study employed (001) and (011) oriented 82$BaTiO_3$-10$BaZrO_3$-8$CaTiO_3$ (BZT–BCT) piezoelectric single crystals, fabricated by the cost-effective solid-state single crystal growth (SSCG) method, in combination with inexpensive, magnetostrictive base metal Nickel (Ni). The off-resonance, direct ME coupling in the prepared Ni/BZT–BCT/Ni laminate composites was found to be strongly dependent on the crystallographic orientation of the BZT–BCT single crystals, as well as the applied magnetic field direction. Larger and anisotropic ME voltage coefficients were observed for the composite made using the (011) oriented BZT–BCT single crystal. The optimized ME coupling of 1 V/cm Oe was obtained from the Ni/(011) BZT–BCT single crystal/Ni composite, in the d_{32} mode of the single crystal, when a magnetic field was applied along its [100] direction. This performance is similar to that reported for the Ni/$Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(Zr,Ti)O_3$ (PMN–PZT) single crystal/Ni, but larger than that obtained from the Ni/$Pb(Zr,Ti)O_3$ ceramic/Ni composites. The results of this work demonstrate that the use of lead-free piezoelectric single crystals with special orientations permits the selection of desired anisotropic properties, enabling the realization of customized ME effects in composites.

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1. Introduction

Magnetoelectric (ME) materials continue to draw a lot of attention due to their wide spectrum of possible applications, including sensing, transduction, memory, and energy harvesting systems [1–4]. Compared to other heterostructures, laminate structured ME composites consisting of magnetostrictive and piezoelectric layers are easier to fabricate and have been found to exhibit superior ME responses [5]. Lead (Pb)-based ferroelectrics such as $Pb(Zr,Ti)O_3$ (PZT), $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT), $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZN-PT), and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(Zr,Ti)O_3$ (PMN–PZT) are often employed in ME composites due to their large piezoelectric coefficients (d_{ij}, g_{ij}) and electromechanical coupling factors (k_{ij}) [6]. Although these ceramics have been found to show good ME performance when combined with magnetostrictive metallic alloys (Terfenol-D, Metglas), because of environmental concerns over the use of toxic and hazardous Pb containing materials, it is now necessary to develop eco-friendly ME composites with lead-free piezoelectric materials. In addition, replacing expensive magnetic alloys with the widely available base metals such as Nickel (Ni) is of interest for the economical production of ME composites.

Most of the investigated lead-free ME composites have been based on $(K,Na)NbO_3$ (KNN), $Na_{0.5}Bi_{0.5}TiO_3$ (NBT), and $BaTiO_3$ (BT). But, due to the low piezoelectricity ($d_{33} < 300$ pC/N, in most cases) of these lead-free ceramics, the corresponding ME composites have been found to exhibit weaker ME responses (on the order of a few hundred mV/cm Oe) than those of the Pb-based ME composites [6]. Recently, a new lead-free ceramic system based on a $Ba(Zr_{0.8}Ti_{0.2})O_3$-$(Ba_{0.7}Ca_{0.3})TiO_3$ solid solution was reported to

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exhibit an elevated piezoelectric coefficient (d_{33} ~620 pC/N) at optimal composition, which is comparable to that of high-end PZT (d_{33} ~500–600 pC/N for PZT-5H) [7,8]. Hence, ME composites prepared using BZT–BCT ceramics are expected to exhibit good ME performance. Nevertheless, there are hardly any reports available on the BZT–BCT ceramic based ME composites [9].

In comparison, single crystal piezoelectric materials demonstrate better piezoelectric performance than their polycrystalline counterparts due to their uniform dipole alignment [10]. Relaxor-based ferroelectric single crystals (PMN-PT, PZN-PT, and PMN–PZT), when cut and poled along specific crystallographic directions, are known to offer excellent strain responses and piezoelectric properties. These rhombohedral structured, domain engineered single crystals display greatly enhanced piezoelectric coefficients and electromechanical coupling factors along the [001] and [011] non-polar orientations, as compared to the [111] spontaneous polarization direction [11]. Results have shown that designing ME composites with special oriented single crystals is an efficient approach to optimizing the ME coupling [12–14]. Interesting relations have been identified between the directional ME coupling and the material constants of the differently oriented piezoelectric single crystals. Although the processing of polycrystalline BZT–BCT ceramics has been considerably well reported, the synthesis of BZT–BCT single crystals has rarely been attempted, since it is expensive as well as challenging to grow lead-free single crystals of usable sizes with uniform composition [15,16]. Recently, single crystals of BZT–BCT with good chemical homogeneity and better piezoelectric properties have been produced via the solid-state conversion of polycrystals into single crystals, using an innovative technique developed by Ceracomp Co., Ltd., Korea [17].

Bulk ME composites are usually developed as trilayered laminates, in which the piezoelectric layer is arranged between two magnetostrictive ones. They are normally operated in the transversely (in-plane direction) magnetized and perpendicularly (thickness direction) polarized mode. This configuration intensifies the strain along the planar direction and minimizes the influence of demagnetizing fields from thickness direction, contributing to a better ME output in low magnetic bias ranges. For a direct ME effect in this mode, because the magnetostrictive phase deforms in the in-plane direction, the piezoelectric phase, due to the interfacial coupling, also has to deform synchronously along the planar direction and induce an electric voltage along the thickness direction [12–14]. This requires the piezoelectric phase to possess a large in-plane strain response (i.e., transverse piezoelectric coefficients, d_{31} and d_{32}) in order to generate a maximized ME output. To exploit this idea further in the case of lead-free piezoelectric single crystals in combination with inexpensive Ni, (001) and (011) oriented $82BaTiO_3$-$10BaZrO_3$-$8CaTiO_3$ (BZT–BCT) single crystals with high transverse piezoelectric coefficients (d_{31} or d_{32}) were chosen for this study. Herein, the off-resonance, direct ME responses of trilayered composites of Ni/(001) BZT–BCT single crystal/Ni, Ni/(011) BZT–BCT single crystal/Ni, Ni/polycrystalline PZT/Ni are compared. The effects of the different orientations of the BZT–BCT single crystals on the ME output was also investigated. The present work is an attempt to develop eco-friendly ME composites with desired sensitivity by replacing the Pb-based piezoelectric ceramics with lead-free ones.

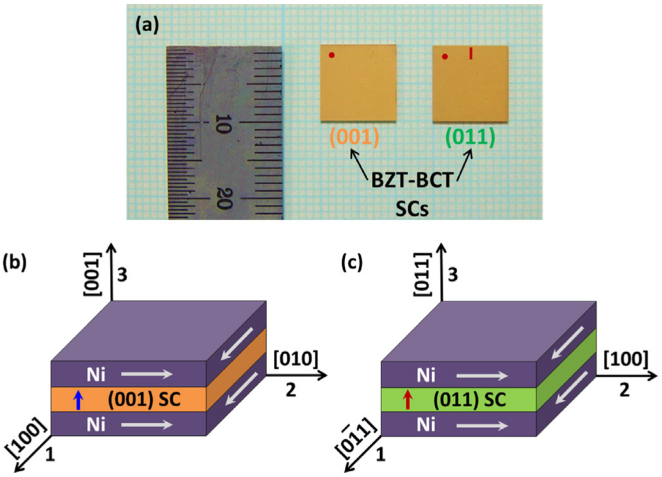

Fig. 1. (a) Photograph of the as-received BZT–BCT single crystals (SCs), (b) and (c) schematic diagrams of the Ni/(001) BZT–BCT/Ni and Ni/(011) BZT–BCT/Ni ME composites, respectively. Here, the arrows on the Ni and BZT–BCT SCs indicate the magnetization and poling directions, respectively.

2. Experimental procedure

ME trilayered composites of Ni/BZT–BCT/Ni and Ni/PZT/Ni were prepared using square (10 mm × 10 mm) plates of Ni (0.25 mm thick), BZT–BCT (0.5 mm thick), and PZT (0.5 mm thick). A photograph of the (001) and (011) oriented BZT–BCT single crystals and the schematics of the Ni/BZT–BCT/Ni laminates are presented in Fig. 1. The properties of the differently oriented BZT–BCT single crystals and the PZT ceramic are summarized in Table 1. High quality single crystals of BZT–BCT which were prepared by the SSCG technique and cut to make their plane vectors parallel to the [001] and [011] directions, were commercially obtained from Ceracomp Co., Ltd., Korea ($LTEX_{Y1}$, $LTEX_{Y2}$). The polycrystalline PZT plate was machined and wire cut from a PZT pellet, which was prepared by pressing and sintering (1250 °C, 2 h) commercially available PZT granules intended for use in high performance piezoelectric sensors and transducers. The X-ray diffraction (XRD, D/Max 2200, Rigaku Corporation, Japan) patterns of the (001) and (011) oriented, rhombohedral $82BaTiO_3$-$10BaZrO_3$-$8CaTiO_3$ single crystals and the polyscrystalline PZT used in this study are shown in Fig. 2. The BZT–BCT single crystals were thickness poled by applying a *dc* electric field of 1.4 kV/mm at room temperature for 10 min. The PZT plate was poled under 4 kV/mm at 120 °C for 20 min, in its thickness direction.

To form the trilayer laminates, magnetostrictive Ni plates (Alfa Aesar, 99.5% metals basis) were bonded to the top and bottom surfaces of the piezoelectric layer using epoxy adhesive (3 M Scotch-Weld^{TM}, DP-460) and cured at 80 °C for 4 h. For the Ni plate, the saturated in-plane magnetostriction (λ_{11}) was measured to be −40 ppm [18]. The in-plane strain-electric field (S–E) behavior of the BZT–BCT and PZT samples was studied using a strain measurement method, which involves a microstrain gauge with Wheatstone bridge. A strain gauge of 5 mm gauge length and 350 Ω nominal gauge resistance (MFLA-5-350-11-1LS, Tokyo Sokki Kenkyujo Co., Ltd, Japan) was mounted on the sample surface with M-bond 200 adhesive. A unipolar electric field (<3 kV/mm) of trian-

Table 1
Dielectric and piezoelectric properties of the BZT–BCT single crystals and the PZT ceramic used in this study.

Material	d_{31} (pC/N)	d_{32}(pC/N)	S^E_{11} (pm^2/N)	S^E_{22} (pm^2/N)	k_{31}	k_{32}	g_{31} (10^{-3} mV/N)	g_{32}(10^{-3} mV/N)
(001)BZT–BCT	−403		44.1		0.41		−19	
(011)BZT–BCT	242	−627	15	47.9	0.55	0.77	17.4	−45
PZT	−112		15.3		0.36		−13.67	

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