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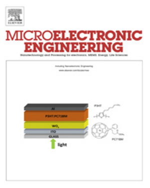

Piezoelectric materials of $(1-x)Pb(Zr_{0.53}Ti_{0.47})O_3$–$xBi(Y_{0.7}Fe_{0.3})O_3$ for energy-harvesting devices

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ABSTRACT

The piezoelectric ceramics $(1-x)Pb(Zr_{0.53}Ti_{0.47})O_3$–$xBi(Y_{0.7}Fe_{0.3})O_3$ [$(1-x)$PZT–xBYF] (where, $x = 0.00$–0.05) has been synthesized by a modified conventional solid state method. Initially the $Pb(Zr_{0.53}Ti_{0.47})O_3$ [PZT] and $Bi(Y_{0.7}Fe_{0.3})O_3$ [BYF] was pre-synthesized and mixed to prepare $(1-x)$PZT–xBYF ceramic composites. The effects of simultaneous addition of BYF on PZT system were measured as a function of sintering temperature, phase formation, microstructure and piezoelectric/dielectric properties. It was found that between 1150 °C and 1190 °C, these ceramics were well sintered showing a maximum density of approximately $\geqslant$97.8% of the theoretical value. X-ray diffraction analysis revealed some minor second phases of $BiYO_3$ and $BiFeO_3$ at $x > 0.01$. A uniform, fine grained and relatively pore-free microstructure is obtained at $x = 0.01$ whereas, the average grain size abruptly decreased on further addition of BYF. All the $(1-x)$PZT–xBYF ceramics doped with various BYF content provided a high T_C in the range of 382–387 °C. It was found that the piezoelectric and the dielectric properties of $(1-x)$PZT–xBYF ceramics vary significantly with increasing BYF content. In addition, the piezoelectric voltage constant (g_{33}), and transduction coefficient ($d_{33} \times g_{33}$) of $(1-x)$PZT–xBYF ceramics have been calculated. For energy-harvesting materials, a high piezoelectric voltage constant expressed by $g_{33} = d_{33}/(\varepsilon_0 \times K_{33}^T)$ (where K_{33}^T is the dielectric constant and ε_0 is dielectric permittivity of free space) is desirable. At the sintering temperature of 1170 °C, the $(1-x)$PZT–xBYF ceramic with 0.01 mol BYF content showed a considerably higher d_{33} and k_p with lower K_{33}^T values which results in significantly higher g_{33} of 53.07×10^{-3} V m/N and ($d_{33} \times g_{33}$) of $20{,}167 \times 10^{-15}$ m^2/N. The large ($d_{33} \times g_{33}$) indicates that the $0.99Pb(Zr_{0.53}Ti_{0.47})O_3$–$0.01Bi(Y_{0.7}Fe_{0.3})O_3$ ceramic is a good candidate material for energy harvesting devices. The detail investigations and observations revealed that, the composition with $x = 0.01$ mol BYF could be the optimum magnitude of doping level in the PZT system.

1. Introduction

Recently, energy-harvesting from the environment has attracted more interest and vibration energy-harvesting is one of methods. A viable family of materials for this purpose is the lead zirconate titanate (PZT) based piezoelectric materials because of their inherent properties. Suitable piezoelectric materials for vibration energy conversion to electrical energy are characterized by a large magnitude of the product of the piezoelectric voltage constant (g_{33}) and the piezoelectric charge constant (d_{33}), which is called the transduction coefficient ($d_{33} \times g_{33}$). According to the definition: $g_{33} = d_{33}/(\varepsilon_0\ K_{33}^T)$ [1–2]. A high transduction coefficient could be obtained from piezoelectric materials with a large d_{33} value and small K_{33}^T. Numerous relevant studies on piezoelectric harvesters have been performed to find such piezoelectric materials in the last decade [3–8]. In particular, a high energy density piezoelectric polycrystalline ceramic composition was reported by Cha et al. with a large transduction coefficient in the $(1-x)Pb(Zr_{0.47}Ti_{0.53})O_3$–$xPb[(Ni_{0.6}Zn_{0.4})_{1/3}Nb_{2/3}]O_3$ system [7]. Lonkar et al. [4] showed a moderately high ($d_{33} \times g_{33}$) in the La substituted $Pb_{0.98}La_{0.02}(NiSb)_{0.05}[(Zr_{0.52}Ti_{0.48})_{0.995}]_{0.95}O_3$ system. Zheng et al. introduced group VIII metal oxides like Fe_2O_3, Co_2O_3 and NiO in the $Pb(Zn_{1/3}Nb_{2/3})_{0.20}(Zr_{0.50}Ti_{0.50})_{0.80}O_3$ system and reported that both ($d_{33} \times g_{33}$) and fracture toughness are improved largely [8]. Recently, we have proposed a method for the composition $(1-x)Pb(Zr_{0.53}Ti_{0.47})O_3$–$xBiYO_3$ ceramics for use in energy harvesting systems [9]. We found that pre-synthesized $BiYO_3$ [BY] was quite effective in lowering the dielectric constant with the retention of good piezoelectric properties and high Curie temperature (T_C). We demonstrated that, BY doped into PZT could remain as a precipitate after sintering and can act as a grain growth

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inhibitor as well as donors in the vicinity of grains. The experimental results indicated that optimum level of doping can significantly improved the ($d_{33} \times g_{33}$). Therefore, for further improvement of ($d_{33} \times g_{33}$), we introduced Fe_2O_3 in this system. Extensive research and development of PZT and PZT based ceramics with Fe_2O_3 have been studied [8,10–12]. Zhu et al. reported that, the dielectric and piezoelectric properties can be tailored which are closely related to the microstructure change, phase evolution, and tetragonal distortion as Fe_2O_3 added in the PZT system [11]. Cross et al. reported that Fe^{3+} incorporate into the PZT lattice and creates defect dipoles, which increase the net polarization [12]. Furthermore, many researchers have been studied PZT based ceramics with bismuth compounds like $BiFeO_3$ [13], $BiAlO_3$ [14] to improve electrical properties with high Curie temperature. Preliminary, we studied the effect of Fe_2O_3 addition on the $0.99Pb[Zr_{0.53}Ti_{0.47}]O_3–0.01Bi[Y_{(1-x)}Fe_x]O_3$ [where, $x = 0.0–0.6$] system [unpublished results]. We found that, the composition with $x = 0.3$ mol of Fe_2O_3 content shows the morphotropic phase boundary region (MPB) and composed of excellent piezoelectric and dielectric properties which also maintained a high ($d_{33} \times g_{33}$). Therefore, in this study, Fe^{+3} was partially substituted for Y^{+3} site in the $BiYO_3$ and $Bi[Y_{0.7}Fe_{0.3}]O_3$ was pre-synthesized as a precursor and subsequently synthesized with PZT.

The main purpose of the present study is to demonstrate the possibility of optimum energy-harvesting material with a high figure of merit ($d_{33} \times g_{33}$), by controlling the pre-synthesized BYF contents. To this end, effects of pre-synthesized BYF content on the crystal structure, microstructure and the piezoelectric/dielectric properties was investigated to search for the optimum $(1-x)$PZT–xBYF compositions with high d_{33} and low K_{33}^{T}.

2. Experimental procedure

A conventional mixed oxide method was used to fabricate $(1-x)Pb(Zr_{0.53}Ti_{0.47})O_3–xBi[Y_{0.7}Fe_{0.3}]O_3$ (where, $0.0 \leqslant x \leqslant 0.05$) [abbreviated as $(1-x)$PZT–xBYF hereafter]. To eliminate the effects associated with the direct co-doping of Fe_2O_3, Bi_2O_3 and Y_2O_3, a pre-synthesis method was used for the fabrication of $(1-x)$PZT–xBYF by a ball mill process. The first stage of the fabrication was the synthesis of $Pb(Zr_{0.53}Ti_{0.47})O_3$ [PZT] and $Bi[Y_{0.7}Fe_{0.3}]O_3$ [BYF]. The stoichiometric amount of the analytical-reagent grade PbO (99.5%, Dansuk, Korea), ZrO_2 (99.5%, Daichi, Japan), TiO_2 (99.9%, High purity Chemicals, Japan) to synthesize PZT; and Bi_2O_3 (99.9%, High purity Chemicals, Japan), Fe_2O_3 (99%, Samchun pure chemical, Korea) and Y_2O_3 (99.9%, High purity Chemicals, Japan) to synthesize BYF were weighed and then ball milled in distilled water using a ZrO_2 media for 24 h. The dried powders were discretely sieved under 100 meshes. The PZT powders were calcined at 850 °C for 2 h and BYF powders were calcined at 800 °C for 2 h in alumina crucibles. The calcination temperature was chosen based on the thermogravimetric and differential scanning calorimetry (TG/DSC-Mettler Toledo) analysis. X-ray diffraction (XRD) analysis at this stage indicated a single phase. The second stage was the fabrication of $(1-x)$PZT–xBYF. The pre-synthesized PZT and BYF powders were weighed and ball-milled for 72 h. The dried powders were pressed as a disk of 15 mm diameter and then cold-isostatically pressed (CIP) under the pressure of 147.2 MPa. The pressed pellets were sintered at various temperatures. To limit PbO loss from pellets, a PbO-rich atmosphere was maintained by placing an equimolar mixture of PbO and ZrO_2 inside a covered alumina crucible. The sintered specimens were then polished to obtain parallel surface and suitable dimension (diameter/thickness $\geqslant$ 15) in order to estimate the piezoelectric properties.

The specimens were characterized by X-ray diffractometer (XRD, Rigaku D/Max-2500H and D8 advance, Bruker) with Cu Kα radiations after calcination and sintering. The morphologies and the microstructures for all of the powders and sintered samples were investigated using a scanning electron microscope (SEM; Hitachi S-2400). In order to measure the electrical properties, silver paste was coated to form electrodes on both sides of the sample, and then subsequently fired at 560 °C for 30 min. For investigating the proper poling condition, each specimen was poled in stirred silicon oil at 120 °C, by applying a dc electric field of 3–6 kV/mm for 45 min and subsequently aged at 120 °C for 3 h. The dielectric and the piezoelectric properties of the aged samples were then measured and evaluated. Preliminary study indicated that the spontaneous polarization was fully saturated under a DC electric field of 4 kV/mm. Therefore, the optimum poling voltage was decided to be 4.0 kV/mm. The standard deviation of the electric properties could exist in the measured samples; therefore, five specimens were prepared and tested for each batch. The piezoelectric coefficient was determined using a d_{33} meter (IACAS; Model ZJ-6B), and the electromechanical, and dielectric properties were calculated by a resonance/anti-resonance measurement method [15] using an impedance/gain phase analyzer (HP-4194A). Temperature dependence of the dielectric constant and the dissipation factor over a range from −25 to 500 °C was measured using an automated system at 1 kHz, whereby HP-4194A and a temperature-control box (−40 to 150 °C: Delta 9023 chamber, 150–500 °C: Lindberg tube furnace) were controlled by a computer system. The temperature was measured using a Keithley 740 thermometer via a K-type thermocouple mounted on the samples. The behavior of the polarization–electric field (P–E) was determined using a Precision LC system (Radiant Technology, Model: 610E).

3. Results and discussions

3.1. Phase analysis of pre-synthesized $Pb(Zr_{0.53}Ti_{0.47})O_3$ and $Bi(Y_{0.7}Fe_{0.3})O_3$

Fig. 1(a and b) shows the XRD patterns for the calcined PZT and BYF, fabricated through conventional mixed oxide process. A fully stabilization of perovskite structure was achieved for the PZT (ICDD No. 01-070-4264) as shown in Fig. 1(a). The calcination temperature was chosen based on TG/DSC. Based on the TG/DSC analysis BYF was calcined at 800 °C for 2 h as shown in Fig. 1(b). The major phase formed for the BYF could be indexed only to the diffraction peak given for $BiYO_3$ (ICDD No. 271047) without any second and/or pyrochlore phases.

3.2. Effects of pre-synthesized BYF content on the microstructure and the densities of $(1-x)$PZT–xBYF ceramics

Fig. 2 shows the SEM images of the $(1-x)$PZT–xBYF ceramics sintered at 1150 °C for 2 h. From the above images, PZT with 0.01 mol of BYF content shows a uniform, fine grained and relatively pore-free microstructure. Whereas, a very small and loosely bonded grains with pores were observed as the BYF content increased to $x > 0.01$. In our previous work we reported that pre-synthesized BY doped into PZT could remain as a precipitate after sintering and BY can act as a grain growth inhibitor [9]. Therefore, in present case the decrease in grain growth with increasing BYF content could be interpreted as the pinning effect of the secondary phase. A detail analysis of BYF doping effects on microstructure and piezoelectric/dielectric properties will be discussed later. The obtained piezoelectric/dielectric and ($d_{33} \times g_{33}$) of $(1-x)$PZT–xBYF ceramics sintered at 1150 °C is shown in Table 1. From the Table 1, it is clear that at $x = 0.01$, $(1-x)$PZT–xBYF specimen sintered at 1150 °C shows a maximum electrical properties with the highest sintering density and well-defined grains. It implies that electric