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High piezoelectric performance and temperature dependence of ferroelectric and piezoelectric properties of $Bi(Mg_{0.5}Zr_{0.5})O_3$–$PbTiO_3$ near morphotropic phase boundary

Longlong Fan[a], Jun Chen[a,b,*], Qi Wang[a], Jinxia Deng[a], Ranbo Yu[a], Xianran Xing[a,b]

[a]Department of Physical Chemistry, University of Science and Technology Beijing, Beijing 100083, China
[b]State Key Laboratory for Advanced Metallurgy, University of Science and Technology Beijing, Beijing 100083, China

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Abstract

According to consideration on the average radius of B-site cation of $BiMeO_3$, we reported that the $Bi(Mg_{0.5}Zr_{0.5})O_3$–$x$$PbTiO_3$ compound at the morphotropic phase boundary (MPB) of $x=0.58$ possesses a piezoelectric coefficient d_{33} as high as 306 pC/N. The optimal piezoelectric and ferroelectric properties near the MPB might be attributed to its lower lattice distortion, as described by change of FWHM value for $\{1\,1\,1\}_{PC}$ peaks. Furthermore, $Bi(Mg_{0.5}Zr_{0.5})O_3$–$x$$PbTiO_3$ displayed stable ferroelectric and piezoelectric properties over a temperature range from ambient temperature to above 160 °C, as exhibited by temperature dependence polarization and strain versus electric field curves and thermal depoling process.

Keywords: Tolerance factor; Ferroelectricity; Piezoelectricity

1. Introduction

Over the past decades, $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) solid solutions around the morphotropic phase boundary (MPB) have been widely utilized as actuator, sensor, and transducer based on high piezoelectric performance [1,2]. In order to achieve better technological properties, many other $PbTiO_3$-based compounds, especially $BiMeO_3$–$PbTiO_3$, have been investigated recently, For $BiMeO_3$-PT, Me can be a cation or a mixture of cations with an valency of +3 [3–10].

In the $BiMeO_3$-PT system, $BiScO_3$-PT (BS-PT) exhibits attractive properties, that it possesses an excellent piezoelectric performance, d_{33} is ~460 pC/N, and a high T_C at MPB, about 450 °C [7,8]. Recently, investigations on Bi(Ni,Me)O_3-PT, Me=Zr or Hf, also exhibit high piezoelectric performance around the MPB position. Their piezoelectric constants, d_{33}, are 400 and 446 pC/N, respectively [9,10]. Except that, other $BiMeO_3$-PT compounds around the MPB exhibit relatively lower d_{33}. It is known that the investigation on high-T_C BS-PT is based on a crystal chemistry tolerance factor (t) approach. And it speculates that a smaller t of $BiMeO_3$ prefers a higher T_C in $BiMeO_3$-PT around MPB. Besides, Grinberg et al. also suggest that the position of MPB is related to the t [11]. So, we can predict the MPB and T_C of the MPB compound in $BiMeO_3$-PT by the t of $BiMeO_3$. Whereas, are the piezoelectric properties related to some crystal structure factors, such as the B-site ionic radius in $BiMeO_3$-PT, t, and et al.? From the references of BS-PT, $Bi(Ni_{0.5}Hf_{0.5})O_3$-PT, and $Bi(Ni_{0.5}Zr_{0.5})O_3$-PT, it can be summarized that these $BiMeO_3$-PT compounds, possessed of excellent piezoelectric properties around MPB, have large average B-site ionic radius in $BiMeO_3$ and similar MPB position. Besides, the substitutions of B-site cations with large ionic radius also exhibit enhanced piezoelectric properties. For instance, in the Zr^{4+} substituted $Bi(Ni_{0.5}Ti_{0.5})O_3$-PT and $Bi(Mg_{0.5}Ti_{0.5})O_3$-PT, their piezoelectric properties have been improved by the substitution of smaller Ti^{4+} by larger Zr^{4+} cation [12–14]. Chen et al. have also summarized parts of reported $BiMeO_3$-PT compound around the MPB, suggesting that when the t of the MPB

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*Corresponding author at: Department of Physical Chemistry, University of Science and Technology Beijing, Beijing 100083, China.
Tel./fax: +86 10 6233 2525.
E-mail address: junchen@ustb.edu.cn (J. Chen).

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composition in $BiMeO_3$-PT accesses that of BS-PT, it will performance a desired piezoelectric coefficient (d_{33}) [15]. Meaning that, when the average ionic radius of the B-site cations in $BiMeO_3$ is approximate to that of Sc^{3+}, the $BiMeO_3$-PT compound may possess a promising piezoelectric property.

Here, we speculate that the co-substitution of Zr^{4+} with other cations, such as Mg^{2+}, in $BiMeO_3$ should exhibit a promising property. Because, the average ionic radius of Zr^{4+} and Mg^{2+} is 0.72, close to that of Sc^{3+}. As is reported previously, the MPB of $Bi(Mg_{0.5}Zr_{0.5})O_3$-xPT (BMZ-xPT) is near at $x=0.55$, which is similar to the BS-PT [16–18]. In this article, we investigated the MPB position of BMZ-xPT again, and found it possesses high piezoelectric performance, $d_{33}=306$ pC/N, which is much larger than the previous work [18]. Also, the ferroelectric and piezoelectric properties as function of both composition and temperature were investigated.

2. Experiment

The objective compounds of $Bi(Mg_{1/2}Zr_{1/2})O_3$–$x$$PbTiO_3$ around the MPB ($0.55 \leq x \leq 0.63$) were synthesized by the conventional solid-state reaction method. Analytically pure Bi_2O_3, PbO, TiO_2, ZrO_2 and $4MgCO_3 \cdot Mg(OH)_2 \cdot 5H_2O$ were homogenized in an agate mortar according to the stoichiometry. The mixed powder was then calcined at 850 °C for 5 h. Calcined powders were pressed into cylindrical compacts at a pressure of 200 MPa, then sintered at 1080 °C for 2 h covered with protective powders of the same composition. The XRD data was collected from the crushed ceramic powders and polished ceramics by the X-ray diffractometer equipped with Cu-$K\alpha$ radiation (X′Pert PRO, PANalytical, Netherlands). For the measurements of electrical properties, the pellets were electroded with silver paste and fired at 550 °C for 30 min. Ferroelectric properties were measured by a ferroelectricity analyzer (TF Analyzer 1000, aixACCT, Germany). The polarization and strain versus electric field ($P(E)$ and $S(E)$) loops at room temperature and the temperature dependence of unipolar $S(E)$ curves were measured at 7 kV/mm with a frequency of 1 Hz. The pellets were first poled in the silicone oil under 7 kV/mm for 10 min. After aging a day, the piezoelectric constant was tested by the piezo-d_{33} meter (ZJ-3, China Academy of Acoustics, China).

3. Results and discussion

It is well known that the MPB position of piezoceramics exhibits optimal piezoelectric and ferroelectric properties. So, we fabricated the compositions of BMZ-xPT with $0.55 \leq x \leq 0.63$ in order to determinate the exact position of MPB. Fig. 1 shows the XRD patterns of BMZ-xPT ceramics and powders. As shown in XRD patterns of powders (Fig. 1a), the MPB of BMZ-xPT system should be around the composition of $x=0.58$. The BMZ-0.63PT compound exhibits obviously tetragonal structure. With decreasing PT content, the compound transfers from tetragonal to rhombohedral phase, as distinguished by the disappearance in the splitting of $\{2\ 0\ 0\}_{PC}$ peak. As x decreases to 0.57, the compound completely transforms to rhombohedral one. It should be noted that the previous reported MPB position was at $x=0.55$ [17,18]. In the ceramics, the coexistence of tetragonal and rhombohedral phases shifts to the composition with higher PT content (Fig. 1b). The similar phenomenon is also observed in La-substituted $BiFeO_3$–$PbTiO_3$ compounds [19]. The internal stress between grains may contribute to that shift. When the ceramic transits from paraelectric phase to ferroelectric tetragonal or rhombohedral one with decreasing temperature, the crystal lattice is distorted, and that will cause internal stress near grain boundaries. The internal stress region may convert the tetragonal to the pseudocubic phase [20]. The zoomed XRD patterns of the $\{2\ 0\ 0\}_{PC}$ peak shows that the coexistence of tetragonal and rhombohedral phases persists over a large range in ceramics. In order to investigate the evolution of lattice distortion, we calculate the FWHM of $\{1\ 1\ 1\}_{PC}$ peak of ceramics and powders after subtracting the $K\alpha_2$ and fitting to pseudo-Voigt profile shape function. Noted that, the change of the FWHM of $\{1\ 1\ 1\}_{PC}$ peak of powders is negligible. While the FWHM of $\{1\ 1\ 1\}_{PC}$ peak of ceramics reaches to the minimum at the position of $x=0.58$ as shown in the inset of Fig. 1b, suggesting the MPB composition possesses lower lattice distortion. The analogical phenomenon is also observed

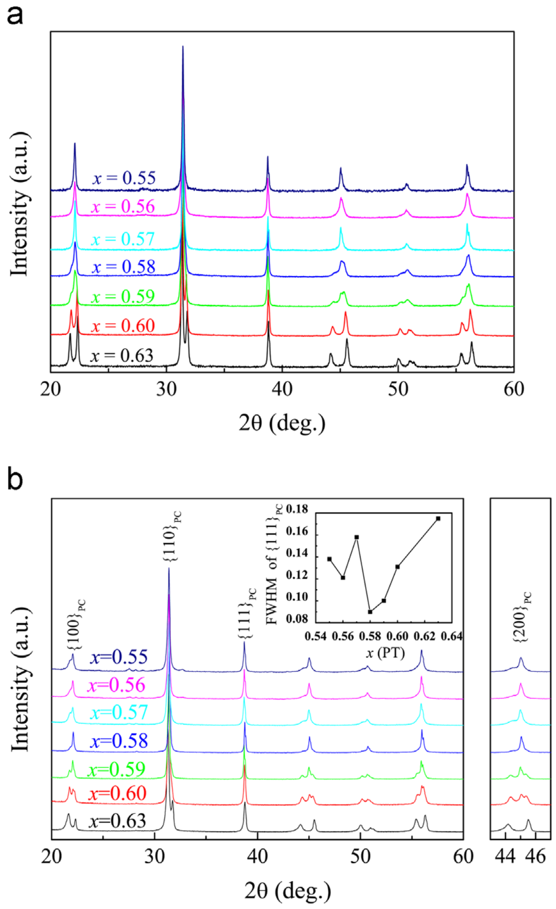

Fig. 1. XRD patterns of (a) powders, and (b) ceramics for BMZ-xPT ($0.55 \leq x \leq 0.63$).