The Study on the Phase Transition and Piezoelectric Properties of $\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3$ - LaMnO_3 Lead-free Piezoelectric Ceramics

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(Received June 21, 2015; Revised July 20, 2015; Accepted July 23, 2015)

ABSTRACT

$\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3$ (BNKT) lead-free piezoelectric ceramics modified by LaMnO_3 (LM) were fabricated by conventional solid-state method. The crystal structure and the morphology of the lead free ceramics were analyzed by XRD (X-ray diffraction) and FE-SEM (Field Emission Scanning Electron Microscopy). The LM modified BNKT ceramics have a phase transition from ferroelectric tetragonal to non-polar pseudo-cubic. Despite decreases in the remnant polarization (P_r) and coercive field (E_c) in the P-E hysteresis loops, the electric-field induced strain properties were significantly enhanced by the LM modification. The highest value of $S_{\text{max}}/E_{\text{max}} = 412 \text{ pm/V}$ at an applied electric field of 5 kV/mm was found in BNKT-0.01LM ceramic.

Key words : BNKT, LaMnO_3 , Phase transition, Actuator, $S_{\text{max}}/E_{\text{max}}$ (normalized strain)

1. Introduction

Piezoelectric materials can convert electrical energy to mechanical energy and are being applied to diversified areas such as ultrasonic sensor, piezoelectric transformer, piezoelectric actuator, resonator, etc. While the piezoelectric actuator application area, in particular, is receiving much interest as information and communication technology is developed among these application areas, materials with a large strain resulting from electric field are advantageous as the piezoelectric material used in piezoelectric actuator modules, and a solid solution of perovskite crystals is suitable for such purpose.¹⁾ Hence, while many studies are being conducted on piezoelectric materials having a high reliability for precise control of devices and excellent strains, Pb-based ceramics represented by $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) are materials with a high piezoelectric constant (d_{33}) and electromechanical coupling coefficient (K_p) and have been applied to many electronic instrument modules including piezoelectric actuator modules for the past few ten years.^{2,3)} However, as regulations against the use of lead is reinforced day by day recently starting with European Union, etc. due to the disadvantages of being toxic for human body and causing environmental contamination since lead composition is contained, studies are being conducted actively on lead-free piezoelectric ceramics to substitute for the lead-based piezoelectric ceramics.⁴⁾ As a representative lead-free piezoelectric material, ferroelectrics such as BaTiO_3 (BT), $(\text{Na,K})\text{NbO}_3$ (NKN)⁵⁾

and materials based on $(\text{Bi,Na})\text{TiO}_3$ (BNT) are mainly used, of which BNT-based materials have excellent characteristics of Remnant Polarization ($P_r = 38 \mu\text{C}/\text{cm}^2$) and a high Curie Temperature ($T_c = 320^\circ\text{C}$)⁶⁾ as compared with BT, etc., making them promising materials⁷⁾ for application as a mobile haptic actuator capable of providing human body with touch sensitivity upon using mobile information instruments as huge deformation occurs upon application of an electric field for a particular composition. However, since the greatest problem of BNT-based materials is a disadvantage that a high voltage should be applied in the poling process of the material due to a high Coercive Electric Field,⁸⁾ studies are being attempted to overcome this by many domestic and foreign study groups where the coercive field is lowered by a method with solid solutions of a BNT-based material being formed with diversified compounds having a perovskite crystal structure such as BaTiO_3 (BT) and $(\text{Bi,K})\text{TiO}_3$ (BKT), etc. Particularly, excellent electrical characteristics among BNT compositions have been reported⁹⁾ when 22 mol% in content of BKT was solid solutioned in BNT-based ceramics since the crystal structure close to a tetragonal system is formed at Morphotropic Phase Boundary (MPB). Therefore, in the present article, $\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3$ (BNKT) composition as a BNT-BKT solid solution has been prepared. Also, studies were conducted on the change in crystal structures and the electrical characteristics of BNKT ceramics by adding about 1 ~ 5 mol% of LaMnO_3 (LM) having a La^{10} -based ABO_3 perovskite structure which has a similar ionic polarizability to that of Bi and a relatively high volatilization temperature because of the possible degradation in electrical characteristics upon firing, considering the low volatilization temperatures of Bi, Na, etc.

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2. Experimental Procedure

In the present article, $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLaMnO}_3$ ($x = 0, 0.01, 0.03, 0.05$) was produced as ceramic specimens through general solid-state reaction method, and effects of LaMnO_3 (LM) on the electrical characteristics of BNKT ceramics were analyzed. After Na_2CO_3 , K_2CO_3 , Bi_2O_3 , TiO_2 , La_2O_3 , and Mn_2O_3 powders of higher than 99% in purity were weighed per composition of each, they were mixed and pulverized for 24 h by ball mill using alcohol as a dispersion medium and zirconia (ZrO_2) balls as a grinding medium. After the powders were dried in a Micro-oven above 100°C for more than 12 h, and subjected to temperature rise in a sintering furnace at $5^\circ\text{C}/\text{min}$, followed by calcination at 800°C for 2 h. About 1 ~ 5 mol% of LM was weighed and added respectively to the calcined BNKT powder, remixed by ball mill for 24 h and dried again in the Micro-oven for more than 12 h, followed by the second calcination process at 800°C for 2 h. $\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-LaMnO}_3$ (to be referred to as BNKT-LM hereafter) powder with calcination completed was placed in a cylindrical mold ($\Phi = 10$ mm), and formed under the pressure of $1 \text{ ton}/\text{cm}^2$. The formed specimens were subjected to a temperature rise at $5^\circ\text{C}/\text{min}$ for heat treatment at 650°C for 4 h, followed by sintering at 1150°C for 2 h. Crystal structures of the ceramic specimens with sintering completed were analyzed using X-ray diffractometer (Rigaku Corporation, D, max 2200V, Japan) and SEM (Jeol, JSM-6700F, Japan). For an analysis of electrical characteristics and dielectric characteristics of the specimen as a function of temperatures, specimens were ground to a thickness of about 1 mm, had the upper and lower part of the specimens coated with silver (Ag) electrode and were heat treated at 700°C for 30 minutes. For the ceramic specimens with heat treatment process completed, dielectric properties as a function of temperatures were measured using an LCR meter (HP 4284A), while P-E hysteresis, electric field induced hysteresis loops as a function

of electric fields were measured for an analysis of electrical characteristics of the specimens using a standard ferroelectric test system (Radiant Technology Co, P-LC-K, USA).

3. Results and Discussion

Figure 1(a) shows a change in the phases of BNKT-LM ceramic specimens produced with a variation in the contents of pure BNKT and LM between 1 ~ 5 mol% by using X-ray diffraction (XRD). As a whole, the specimens appeared to exhibit a perovskite crystal structure, and appropriate solid solutioning in BNKT ceramics is considered to have occurred in the case of adding 1,3 mol% of LM since no second phase was observed except for the specimen with addition of 5 mol% of LM. However, there was an excessive change in the amount of ions in the case of 5 mol% of LM added suggesting appearance of the secondary phase. Also, the overall XRD pattern was moved increasingly to the left, the higher the LM contents. This suggests that the XRD pattern was moved to a low angle according to Bragg's law as the size of overall crystal structure of the BNKT specimen with LM added was increased in comparison with the pure BNKT since the ionic radius of La ($1.06 \text{ \AA}$) was larger than that of Bi ($1.03 \text{ \AA}$).¹¹⁾ Also, to analyze effects of LM contents on the crystal structures of a specimen more closely, the XRD pattern near $35^\circ \sim 50^\circ$ is shown in Fig. 1(b). Whereas detection of separation of (002)/(200) XRD peaks indicating a tetragonal system at MPB near 46° may be affirmed in the case of the pure BNKT,¹²⁾ phase transition to a Pseudo-Cubic structure may be seen to have occurred as the (002)/(200) X-ray diffraction peaks appearing in the XRD pattern for the pure BNKT were changed to one peak with addition of LM.¹²⁾ Through such phenomenon, it may be inferred that LM not only caused a change in crystal structures for the BNKT specimen but also had profound effects on a change in electrical characteristics.

Surface microstructures of the pure BNKT and the

Fig. 1. The XRD pattern of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLM}$ ($x = 0, 0.01, 0.03, 0.05$) ceramics sintered at 1150°C : (a) The XRD pattern of $20^\circ \sim 70^\circ$, (b) The XRD pattern of $35^\circ \sim 50^\circ$.

ceramic specimens produced with a variation in the LM contents between 1 ~ 5 mol%, respectively, are shown in Fig. 2. While each specimen is affirmed to have a dense microstructure as a whole, particle sizes were shown to be increased with LM contents. Namely, while the average particle size calculated through linear intersection method was about 0.9 μm in the case of pure BNKT without addition of LM as shown in Fig. 1(a), the average particle size was increased to about 1.5 μm with an increase in the added amounts of LM in the case of a specimen with addition of 5 mol% as shown in Fig. 1(d). Such result is similar to the study results for PLZT with La doped in A-site,¹³⁾ and appears to be a result of promoting interdiffusion as magnetic grid defects due to imbalance in electric charges are produced locally while trivalent La ions with an ionic radius different from those of Bi, Na ions are substituted in divalent A-sites and trivalent Mn(0.645 Å) ions with an ionic radius different from that of Ti(0.605 Å) ion are solid solutioned in tetravalent B-sites also.^{13,14)}

Figure 3 shows P-E characteristic curves (Hysteresis loops) for an analysis of ferroelectric and electrical characteristics of individual specimens. As Remnant Polarization(P_r) of the pure BNKT specimen was measured to be 38.7 $\mu\text{C}/\text{cm}^2$ and Coercive Electric Field(E_c) about 2.59 kV/mm, affirming to show typical ferroelectric characteristics. However, as LM was added, the phenomenon appeared where the remnant polarization and the coercive electric field were decreased, and the so-called Relaxorferroelectric characteristics were affirmed to appear where the remnant polarization was relatively decreased although the maximum polarization for the specimens with 1 mol% of LM added was measured to be similar to that of the pure BNKT at 46.7 $\mu\text{C}/\text{cm}^2$.¹⁵⁾ However, for the specimens with addition

of 3, 5 mol%, the maximum polarization and the remnant polarization characteristics were drastically degraded so as to be shown close to paraelectric characteristics which form a linear curve as a function of the applied electric field. According to the analysis results of XRD patterns shown in Fig. 1, such change is considered attributable to the change of dipoles within the crystal structure from a tetrahedral structure with polarity to a pseudo-cubic structure with no polarity due to grid distortion within the specimen as La and Mn ions of mutually different ionic radii are solid solutioned in A-site and Bi-site. Ferroelectric characteristics of the specimen appears to have been changed to Relaxorferroelectric characteristics, and then to paraelectric characteristics, as more grid distortion phenomenon occurred with a gradual increase in the solid solution contents of LM.

In Fig. 4(a), characteristic curves of bipolar Electric Field Induced Strain are shown for the specimens of BNKT and BNKT with LM added. As analyzed in Fig. 3, specimens of the pure BNKT exhibited a ferroelectric hysteresis loop of Butterfly Shape, Negative Strain(S_{neg}) appearing as the result of reversal in the direction of ferroelectric domain inside BNKT according to the applied electric field when the electric field reached the coercive electric field can be seen to be as large as about -0.12%. Based on such hysteresis loops, existence of a ferroelectric domain in the BNKT specimen can be proved.¹⁴⁾ However, the negative strain (S_{neg}) exhibiting ferroelectric characteristics was greatly decreased as LM was added according to the contents of each, and the maximum strain(S_{max}) was shown to be about 0.25% when an electric field of 5 kV/mm was applied to the specimen with 1 mol% of LM added. Such change in electric field induced strains may be predicted as a phenomenon where the electric field induced strain is drastically increased as a phase transition back to the ferroelectric phase occurs in the specimen by application of an electric field to the specimen where phase transition occurred from a ferroelectric phase to a nonpolar phase without polarity due to addition of LM. According to the study results by research groups including

Fig. 2. The surface microstructure of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-x\text{LM}$ ($x=0, 0.01, 0.03, 0.05$) ceramics sintered at 1150°C: (a) Pure BNKT ($x=0$), (b) $0.99\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-0.01\text{LM}$ ($x=0.01$), (c) $0.97\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-0.03\text{LM}$ ($x=0.03$), (d) $0.95\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-0.05\text{LM}$ ($x=0.05$).

Fig. 3. The hysteresis loops of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-x\text{LM}$ ($x=0, 0.01, 0.03, 0.05$) ceramics.

Fig. 4. (a) The electric field induced strain curves of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLM}$ ($x=0, 0.01, 0.03, 0.05$) ceramics, (b) The unipolar electric field induced strain curves of pure BNKT ($x=0$) and BNKT-0.01LM ($x=0.01$) ceramics.

Rödel of Germany,¹⁷⁾ phase transition has been observed as a result of real-time analysis of BNT-based ceramic specimens macroscopically exhibiting no polarity by synchrotron X-ray diffraction method,^{17,18)} etc. while an actual electric field was being applied.

Figure 4(b) shows a characteristic curve of unipolar electric field induced strain for the pure BNKT and the most excellent BNKT-0.01LM with excellent induced strain characteristics among the characteristic curves of bipolar electric field induced strain shown in Fig. 4(a). In the case of piezoelectric actuators, the induced strain characteristics are used mainly under an unipolar electric field rather than a bipolar electric field, because proportional control is easier under the unipolar electric field. For the pure BNKT specimen, the maximum strain is affirmed to be about 0.17% under the applied electric field of 5 kV/mm, while the maximum strain for the BNKT specimen with 1 mol% of LM added was increased to about 0.26%. Based on this, Normalized Strains ($S_{\text{max}}/E_{\text{max}}$) were calculated for the pure BNKT specimen and the BNKT specimen with 1 mol% of LM added. In the case of pure BNKT, it was calculated to be about 237 pm/V, while a considerably high value of about 412 pm/V was observed for the specimen with LM added. Since a considerably high value for the electric field induced strain was exhibited in comparison with the pure BNKT, a very high promise is indicated when employed as actuator modules applied across the entire industry.

In Fig. 5, dielectric constants (Fig. 5(a)) and dielectric losses (Fig. 5(b)) are shown as a function of temperature of the specimens. When dielectric losses of Fig 5(b) are analyzed, the dielectric losses of all specimens show low values on the level of about 5~7% at room temperature, while the dielectric loss values at a high temperature were shown to be decreased more than at room temperature. Also, a tendency was observed where loss values were increased and then decreased with an increase in temperature. This tem-

Fig. 5. The temperature dependent dielectric constant and dielectric loss of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLM}$ ($x=0, 0.01, 0.03, 0.05$) ceramics: (a) dielectric constant of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLM}$ ($x=0, 0.01, 0.03, 0.05$), (b) dielectric loss of $(1-x)\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3\text{-xLM}$ ($x=0, 0.01, 0.03, 0.05$).

perature section is the so-called $T_{\text{F-R}}$ (ferroelectric-to-relaxor transition temperature) section,¹⁹⁾ and a phenomenon was observed where the $T_{\text{F-R}}$ section was extensively diffused and moved toward a low temperature, the higher the LM contents, whereas the $T_{\text{F-R}}$ section was formed near 135°C in the case of pure BNKT. As analyzed in XRD patterns, P-E hysteresis characteristics and characteristic curves of electric field induced strain, ferroelectric characteristics are degraded in this phenomenon, because of which the $T_{\text{F-R}}$ section is moved to a lower temperature by lowering the long range ferroelectric order of the pure BNKT as a result when the phase transition occurs from a tetragonal structure showing ferroelectric phase to a pseudo-cubic structure showing nonpolar phase with addition of LM to BNKT according to the XRD pattern. Similar phenomenon to this one has been reported when SrZrO_3 (SZ) was added to BNT base.²⁰⁾

4. Conclusions

In the present study, according to the results of producing the pure BNKT specimen and the specimens with 1 ~ 5 mol% of LM added to the basis of $\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3$ composition through solid-state reaction method and analyzing the crystal structures and the electrical characteristics, improvement in electrical characteristics was realized for application to piezoelectric actuator modules. According to the results of observation through FE-SEM of the microstructures of lead-free piezoelectric ceramics produced in the present study, proper densification is affirmed to have been realized as a whole. Crystal structures were analyzed using XRD, where addition of LM caused transition in a crystal structure of the specimen to a pseudo cubic structure with nonpolar characteristics while causing solid solutioning of La and Mn ions having mutually different ionic radii, although the pure BNKT was affirmed to have a tetragonal system structure exhibiting ferroelectric characteristics. In terms of a change in the P-E hysteresis loop characteristics, a change from the ferroelectric characteristics to the Relaxorferroelectric characteristics with a decrease in coercive electric field and remnant polarization were affirmed to have occurred as LM was added. Particularly, in the case of specimens with 1 mol% of LM added, it was observed that the negative electric field induced strain showing coercive electric field and ferroelectric domain was markedly decreased and the maximum induced strain was most excellent. Also, according to the results of showing characteristic curve of unipolar electric field induced strain and $S_{\text{max}}/E_{\text{max}}$ values digitizing strains based on this curve, it is predicted that high electrical characteristics as compared with applied voltages may be expected upon application to piezoelectric actuator modules inasmuch as $S_{\text{max}}/E_{\text{max}}$ showed a considerably high value of about 412 pm/V at an applied electric field of 5 kV/mm when compared with the value for the pure BNKT.

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