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Microstructure and electrical properties of $(Na_{0.5}K_{0.5})_{1-2x}Mg_xNbO_3$–$Bi_{0.5}Na_{0.5}TiO_3$ lead-free piezoelectric ceramics

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Abstract

0.975[$(Na_{0.5}K_{0.5})_{1-2x}Mg_xNbO_3$]–0.025($Bi_{0.5}Na_{0.5}TiO_3$) (KNMN–BNT, $x=0$, 0.01, 0.02, 0.03, 0.04 and 0.05) lead-free piezoelectric ceramics were fabricated by the conventional solid-state sintering method. The dependence of Mg content on the microstructure and electrical properties of the ceramics is investigated. The X-ray diffraction (XRD) analysis revealed that an appropriate amount of Mg diffused into the KNN–BNT lattice to form a stable solid solution, the ceramics possessed a pure perovskite structure, and a morphotropic phase boundary (MPB) between the orthorhombic and tetragonal phases was observed with the composition of $0.02 \le x \le 0.05$. The orthorhombic–tetragonal transition temperature (T_{O-T}) is less than 95 °C and the Curie temperature (T_c) is almost unchanged (~360 °C) with the increase of MgO content. The ceramics with $x=0.02$ showed enhanced piezoelectric and ferroelectric properties because of close proximity to the MPB, i.e., d_{33}~210 pC/N, k_p~0.41, $2E_c$~22.4 kV/cm and $2P_r$~39.2 μC/cm^2. Moreover, the dielectric properties exhibited optimal effects with $x=0.02$, that is ε_r~637 and tan δ~0.09. These results indicate that the introduction of MgO is an effective method to improve the density as well as the electrical properties and the temperature stability of the KNN–BNT ceramics. As a result, the KNMN–BNT ceramic is a promising candidate for lead-free piezoelectric materials.

Keywords: C. Electrical properties; Lead-free ceramics; $(Na_{0.5}K_{0.5})NbO_3$; Piezoelectricity

1. Introduction

Lead-based ceramics have been widely used for various ferroelectric and piezoelectric applications, such as sensors, capacitors and actuators [1,2]. However, the lead-based materials have resulted in serious environmental pollution and human health problem during material preparation and waste of products. Therefore, they will be banned in future in the view of environment protection. In order to solve this problem, lead-free ceramics with excellent electrical properties have been attended extensively for replacing these lead-based ceramics [3–16].

$K_{0.5}Na_{0.5}NbO_3$ (KNN) ceramic is one kind of candidates in recent years due to its high piezoelectric properties, high Curie temperature (T_c) and environmental friendliness [3–6,14–18]. However, it is also difficult to process into dense ceramics using the conventional sintering method due to the high volatility of alkaline elements at elevated temperatures [6,7,15,19]. In order to solve these problems to obtain well-sintered and dense KNN materials with excellent electrical performance, some ABO_3-type compounds have been doped into KNN. Among these, a small amount of $Bi_{0.5}Na_{0.5}TiO_3$ (BNT) ceramics has been added into KNN to form KNN-based solid solutions for improving sintering behavior [7–9,12–14,16,19]. It was reported that KNN–BNT ceramics with about 2–3 mol% BNT could form a tetragonal phase near the morphotropic phase boundary (MPB) region and exhibit excellent electromechanical properties [9]. Moreover, it was also found that the KNN-based ceramics could be well-sintered and the density could be effectively improved by adding Mg^{2+} into KNN-based materials [20,21]. However, there are few reports on the microstructure and electrical properties of Mg^{2+}-modified KNN–BNT ceramics to date.

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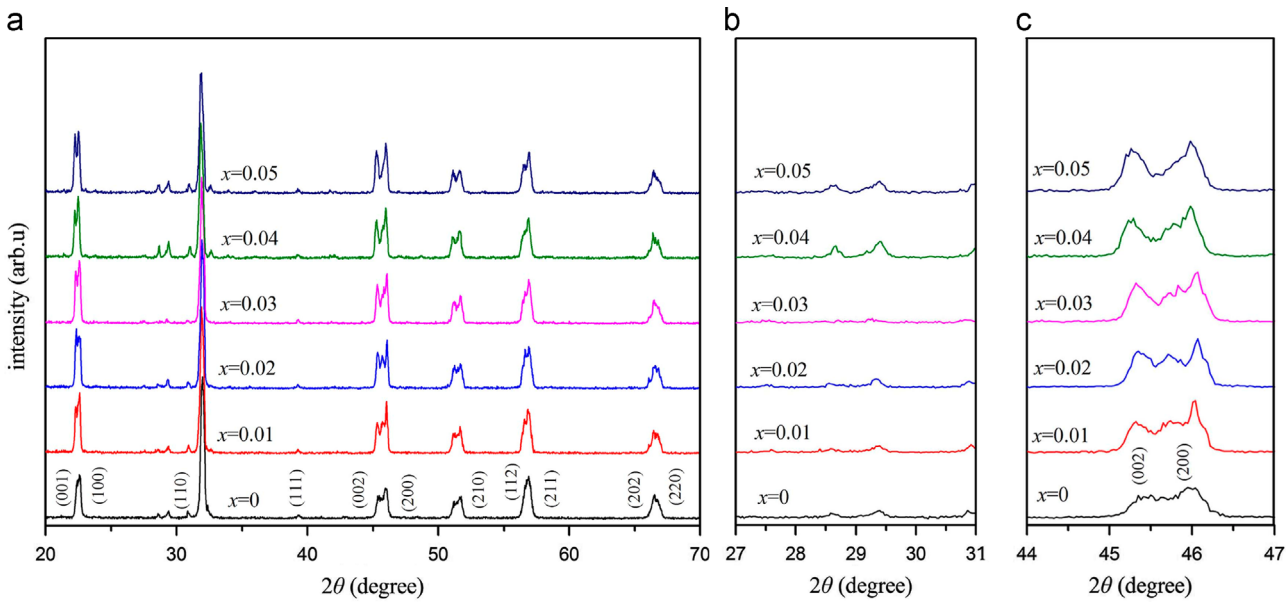

Fig. 1. XRD patterns of KNMN–BNT ceramics. (a) θ=20–70°, (b) θ=27–31° and (c) θ=44–47°.

In the present work, $0.975[(Na_{0.5}K_{0.5})_{1-2x}Mg_xNbO_3]$–0.025 ($Bi_{0.5}Na_{0.5}TiO_3$) (abbreviated as KNMN–BNT) piezoelectric ceramics were fabricated by the conventional solid-state method. The effect of Mg content on the microstructure and electrical properties of KNMN–BNT ceramics was investigated, and the underlying physical mechanism was also illuminated.

2. Experimental procedure

The $0.975[(Na_{0.5}K_{0.5})_{1-2x}Mg_xNbO_3]$–$0.025(Bi_{0.5}Na_{0.5}TiO_3)$ (x=0, 0.01, 0.02, 0.03, 0.04, and 0.05) ceramics were prepared by the conventional solid-state reaction method. Raw materials of the present work were Na_2CO_3 (99.8%), K_2CO_3 (99.8%), Nb_2O_5 (99.5%), Bi_2O_3 (99.0%), TiO_2 (99.99%) and MgO (99.0%), and then the powders were ball milled with ZrO_2 balls for 24 h using ethanol as the medium. Homogenous mixtures were dried and calcined at 850 °C for 6 h. The calcined powders were reground and mixed with 5 wt% polyvinyl alcohol (PVA) solution as a binder, and then pressed into pellets of ~10 mm in diameter and ~1.0 mm in thickness by a uniaxial pressure of 10 MPa. After burning off PVA, these ceramic disks were sintered in the temperature range of 1070–1120 °C for 3 h in air. Silver pastes were fired at ~700 °C for 10 min on both sides as electrodes for electrical measurement. All samples were polarized in a silicon oil bath at room temperature under a direct current electric field of ~3.5 kV/mm for 20 min.

The phase structure of these specimens was examined by using an X-ray diffraction (XRD) in the θ–2θ scan mode (DX1000, PR China). Surface morphologies of these specimens were determined by scanning electron microscopy (SEM) (Philips, XL30). The density of the sintered ceramics was measured by the Archimedes method. The dielectric behavior as a function of the measurement temperature of these samples was examined by using a programmable furnace with an LCR analyzer (HP 4980, Agilent, USA). The ferroelectric properties of these specimens were measured by using the Radiant precise workstation (Radiant Technologies, Medina, NY). The piezoelectric constant of these ceramics (d_{33}) was studied using a piezo-d_{33} meter (ZJ-3A, China).

3. Results and discussion

Fig. 1(a) shows the X-ray diffraction (XRD) patterns of KNMN–BNT ceramics as a function of Mg content, measured at room temperature and in the 2θ range of 20–70°. It can be seen that the main phase of all the ceramics exhibits a perovskite structure. Fig. 1(b) shows the expanded XRD micrographs of KNMN–BNT ceramics, measured at room temperature and in the 2θ range of 27–31°. This result indicates that the ceramics is almost in pure perovskite phase and most Mg^{2+} have diffused into the KNMN–BNT ceramics to form a new stable solid solution when $x \leq 0.03$. However, there exists a small amount of secondary phase when $0.04 \leq x \leq 0.05$, which could be attributed to the introduction of excessive MgO [21]. To further study the effect of Mg content on the phase structure of KNMN–BNT ceramics, the expanded XRD patterns of all ceramics have been performed in the 2θ of 44–47°. Fig. 1(c) shows the expanded XRD micrographs of KNMN–BNT ceramics, measured at room temperature. It is clear that the KNMN–BNT ceramics exhibit tetragonal symmetry at room temperature when $x < 0.02$. With the increase of Mg content, the phase structure of the ceramics transforms from tetragonal phase to a coexistence of orthorhombic and tetragonal phase for $0.02 \leq x \leq 0.05$. These characteristics suggest that the KNMN–BNT ceramics has an orthorhombic–tetragonal morphotropic phase boundary (MPB) in this region, which is expected to display good piezoelectric properties [9,12,19].

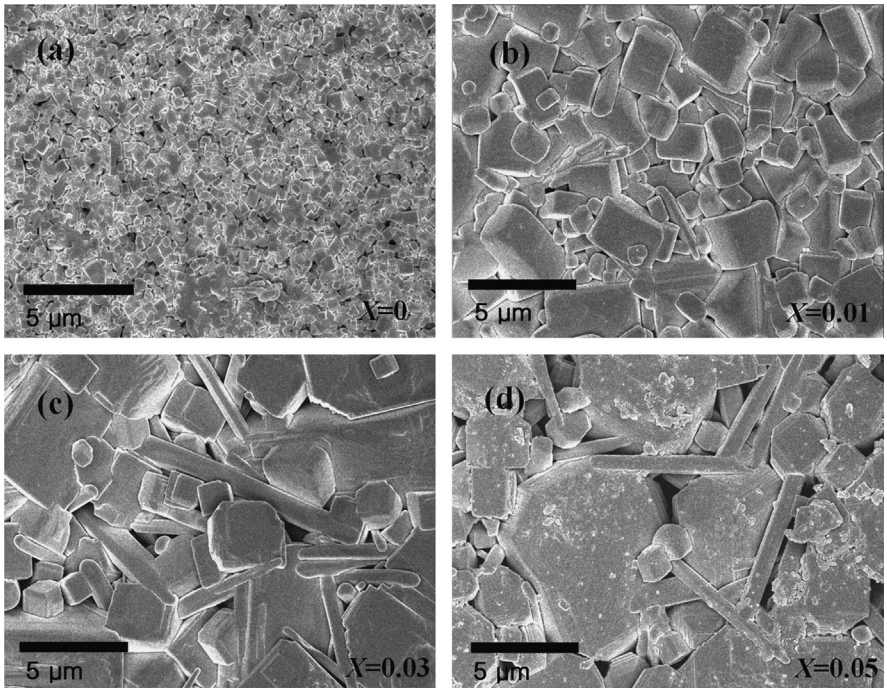

Fig. 2. SEM surface micrographs of KNMN–BNT ceramics with different Mg contents, (a) $x=0$, (b) $x=0.01$, (c) $x=0.03$, and (d) $x=0.05$.

Fig. 2(a), (b), (c) and (d) shows the SEM patterns of surface morphologies of KNMN–BNT ceramics with $x=0$, 0.01, 0.03 and 0.05, respectively. It was observed that the Mg content strongly affects the microstructure of the KNMN–BNT ceramics. With the increase of Mg content, the KNMN–BNT ceramics become much denser, and the average grain size increases. This may be attributed to the introduction of Mg^{2+} ions. The partly substitution of Mg^{2+} into the Nb^{5+} and Ti^{4+} sites could result in the generation of more oxygen vacancies. These oxygen vacancies could help the mass transport during sintering process and as a result, acts in helping to improve the grain growth of the KNMN–BNT material system [21]. In order to further characterize the effect of Mg content on the density of all ceramics, the densities have been measured as a function of Mg content, as shown in Fig. 3. It was observed that the relative densities of the ceramics are all above 93.5%. This result indicates that all the ceramics are well-sintered in this work. The measured density of these ceramics is in the range of 4.32–4.38 g/cm^3, and which slowly increases with increasing Mg content. The relative density also increases and reaches the maximum (about 97%) when $x=0.05$. This phenomenon could also be attributed to the introduction of Mg^{2+} ions. As the Mg^{2+} ($r_{Mg^{2+}} \sim 0.72$ Å) partly replaces the K^+ ($r_{K^+} \sim 1.64$ Å) and Na^+ ($r_{Na^+} \sim 1.39$ Å) sites in KNN, the lattice distorts and the KNMN–BNT ceramics actually become denser [5,22]. Therefore, the introduction of MgO plays an important role in the improvement of the densification of KNMN–BNT ceramics.

The temperature dependence of the dielectric constant (ε_r) of KNMN–BNT ceramics (x=0, 0.01, 0.02, 0.03, 0.04, and 0.05) measured at 10 kHz is shown in Fig. 4(a). It is found that the KNMN–BNT ceramics undergo two phase transitions, which include from orthorhombic to tetragonal (T_{O-T}) and from tetragonal to cubic (T_c). The highest ε_r value of KNMN–BNT ceramics at T_c appears when $x=0.02$, which could be explained on the basis of the mixed structure nature at this MPB composition [23,24]. This is consistent with the XRD examined result (as shown in Fig. 1). It can be seen that the T_c is ~352 °C and the T_{O-T} is below room temperature and so it cannot be observed when $x=0$, which indicates that the ceramics expressed the tetragonal structure at room temperature when $x=0$. This is also in agreement with the XRD result. The variations of T_c and T_{O-T} as a function of Mg content are summarized in Fig. 5. The T_c value firstly increases with an increase of Mg content from 0 to 0.01 (~352 °C), and then remains almost unchanged with the Mg content from 0.01 to 0.05 (~365 °C). The T_{O-T} value firstly increases when $0 \le x \le 0.01$, and then is almost unchanged when $0.01 \le x \le 0.04$, and at last decreases slowly when $0.04 \le x \le 0.05$. As a result, the T_{O-T} value of these ceramics is in the range from room temperature to ~95 °C. These phenomena may be attributed to the introduction of Mg^{2+} ions, resulting in complex occupations of the A and B sites in an ABO_3 perovskite structure [19]. Fig. 4(b) shows the temperature dependence of the dielectric loss for KNMN–BNT ceramics with $x=0$, 0.01, 0.02, 0.03, 0.04 and 0.05, measured at 10 kHz. The sample with $x=0.02$ shows a lower and more stable dielectric loss characteristics, when the tan δ is lower than 0.03 and almost keeps constant in the temperature range from room temperature to 340 °C.

Fig. 6 shows the dielectric constant (ε_r) and dielectric loss (tan δ) of KNMN–BNT ceramics as a function of Mg content, measured at room temperature and 100 KHz. The ε_r value decreases gradually with the increase of Mg content. It is

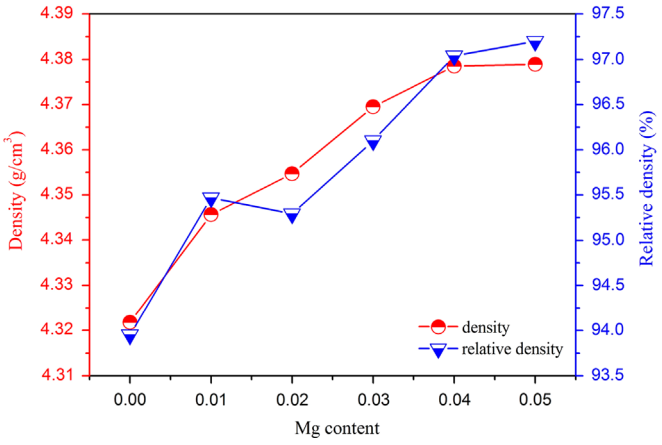

Fig. 3. Measured and relative density of KNMN–BNT ceramics with different Mg contents.

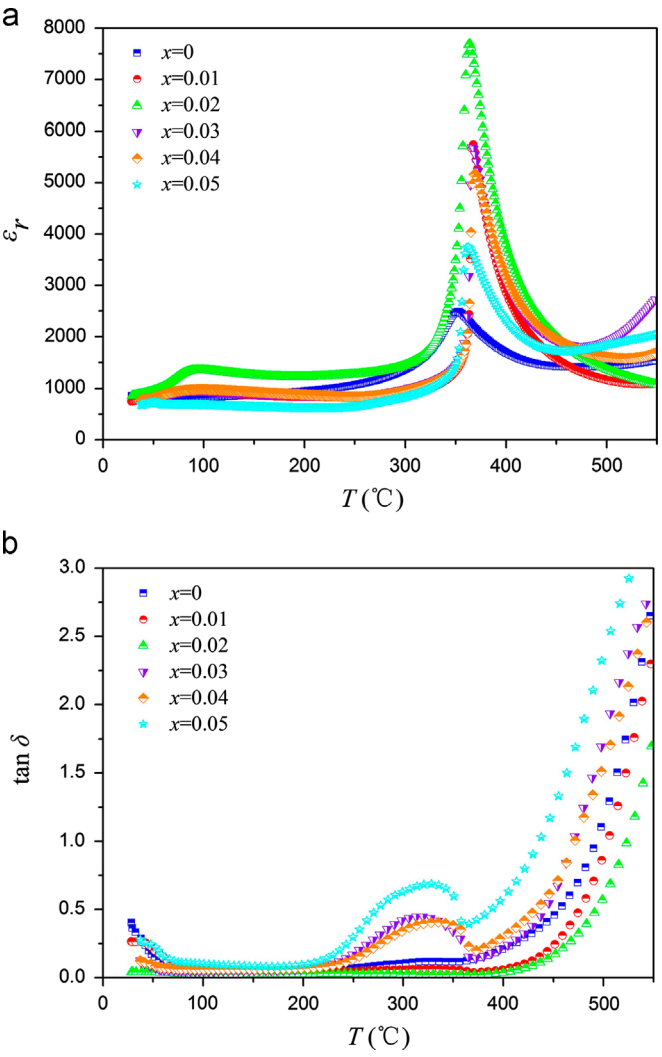

Fig. 4. Temperature dependence of (a) dielectric constant and (b) dielectric loss of KNMN–BNT ceramics.

found that ε_r value of all the ceramics varies from 530 to 735. The tan δ value firstly decreases, and reaches the minimum at $x=0.01$, and then increases slowly with further increasing of

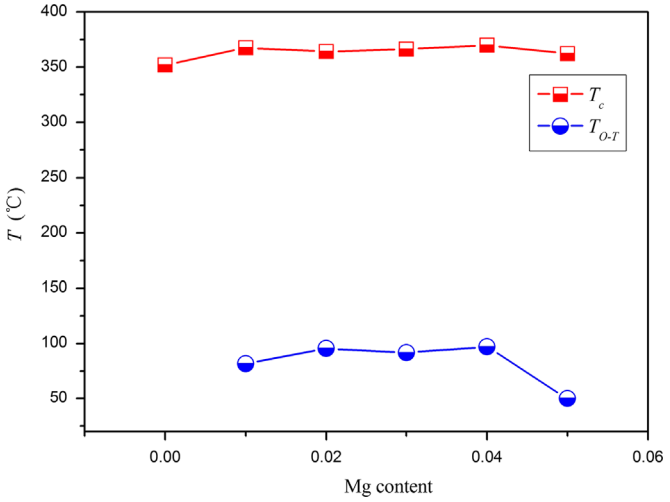

Fig. 5. T_c and T_{O-T} values of KNMN–BNT ceramics as a function of Mg content.

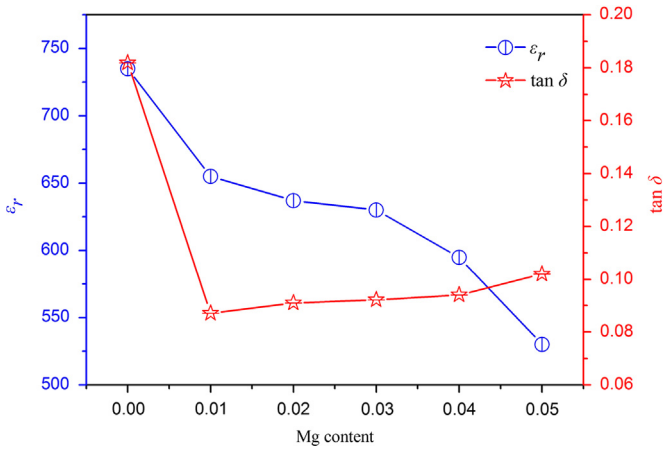

Fig. 6. Dielectric constant and dielectric loss of KNMN–BNT ceramics as a function of Mg content.

Mg content. As a result, the ceramics have the lowest tan δ of about 0.09 with $x=0.01$ and all the tan δ values vary less than 0.19. It can be seen that the tan δ value decreases obviously after introducing MgO comparing with pure KNN–BNT ceramics. It is well-known that the dielectric properties (ε_r and tan δ) of KNN ceramics are related to the density and grain-size distribution [19,25]. It can be seen from Fig. 3 that the densities of the KNMN–BNT ceramics are improved by the addition of Mg content. Therefore, the change of ε_r and tan δ could be attributed to the increase of density of KNMN–BNT ceramics with increasing Mg content. According to Fig. 3, the maximum value of density is ~4.38 g/cm^3 when $x=0.05$, which is consistent with the minimum value of ε_r and the relatively higher value of tan δ when $x=0.05$. Moreover, it can be found from Fig. 5 that the dielectric properties change faster when $x \geq 0.04$ than $0.01 \leq x \leq 0.04$, which may be due to the increase of electrical conductivity and leakage current, as a result, the dissipation factor of KNMN–BNT ceramics increases obviously with the increase of Mg content when $x \geq 0.04$ [26].

Fig. 7 shows the polarization versus electric field (*P–E*) hysteresis loops of KNMN–BNT ceramics as a function of Mg content, measured at 10 Hz and a room temperature of ~20 °C. It is observed that the KNMN–BNT ceramics exist in typical hysteresis loops. The *P–E* loop shows a large leakage current when $x=0$, and this phenomenon may be attributed to the lower density of the ceramic without doping MgO (as shown in Fig. 3) that results in a large leakage current. With the change of Mg content in $(Ka_{0.5}Na_{0.5})_{1-2x}Mg_xNbO_3$–$Bi_{0.5}Na_{0.5}TiO_3$ ceramics from $x=0.01$ to $x=0.02$, the $2E_c$ value decreased and the $2P_r$ value increased, and then expressed a lower $2E_c$ value of ~22.4 kV/cm and a larger $2P_r$ value of ~39.2 μC/cm^2 when $x=0.02$. However, with further increasing Mg content from 0.03 to 0.05, the change of $2E_c$ and $2P_r$ values showed an opposite trend comparing with $0 \le x \le 0.02$. As a result, the KNMN–BNT ceramics have a better ferroelectric behavior when $x=0.02$. It is known that the oxygen vacancies are restraining the reversal of the spontaneous polarization of ferroelectric domains under an applied electrical field, leading to a decrease in P_r and increase in E_c [27,28]. Therefore, the difference in ferroelectric properties with different Mg contents should be contributed to the deviation of Ti^{4+} and Nb^{5+} from the center of the oxygen octahedrons because of the introduction of Mg^{2+} ions. Moreover, E_c reaches the lower value and P_r reaches the higher value when x=0.02, which could be attributed to the formation of MPB, resulting in more possible polarization states. This could also be demonstrated in Fig. 1.

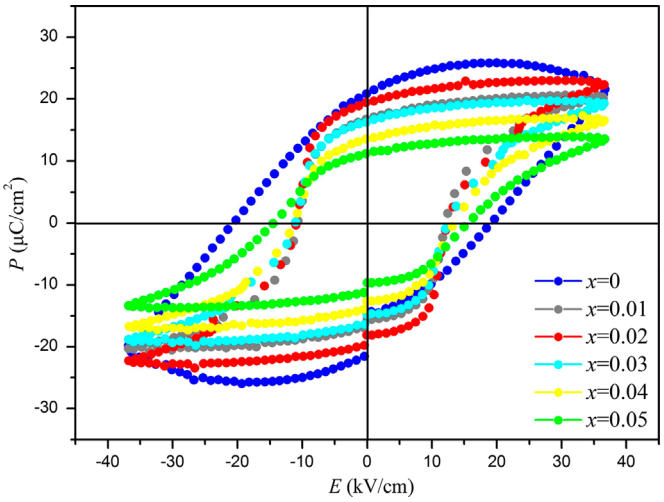

Fig. 7. *P–E* loops of KNMN–BNT ceramics as a function of Mg content.

Fig. 8 shows the piezoelectric constant (d_{33}), the planar electromechanical coupling factor (k_p), and the mechanical quality factor (Q_m) of the KNMN–BNT ceramics as a function of Mg content. As shown in Fig. 8, the d_{33} value enhances with increasing Mg content when $x \le 0.02$, and reaches a maximum value of ~210 pC/N at $x=0.02$, and then decreases with further increasing Mg content. Similar to the change of d_{33} values, the k_p value of KNMN–BNT ceramics also reaches a maximum value of ~0.41 at $x=0.02$. However, a low Q_m value is demonstrated in the range of 45–86. These results confirmed that the KNMN–BNT ceramics demonstrate the highest piezoelectric behavior of d_{33} and k_p at room temperature when $x=0.02$ because of the involvement of a MPB. It is well-known that the improvement of piezoelectric properties of the ceramics near the MPB could be mainly attributed to the increase of spontaneous polarization vectors resulting from the coexistence of two phases [12,19]. In the present work, the ceramic with x=0.02 exhibits a coexistence of two phases, and hence possesses more possible polarization vectors. Therefore, the highest piezoelectric properties of d_{33}~210 pC/N and k_p~0.41 have been demonstrated for the KNMN–BNT ceramics with x=0.02. Moreover, it is known that a lower E_c value results in a more efficient poling of ceramics, and a higher P_r value benefits the piezoelectricity [28]. As shown in Fig. 7, the KNMN–BNT ceramics expressed a better ferroelectric property when $x=0.02$, as a result, which could benefit to the improvement of d_{33} value.

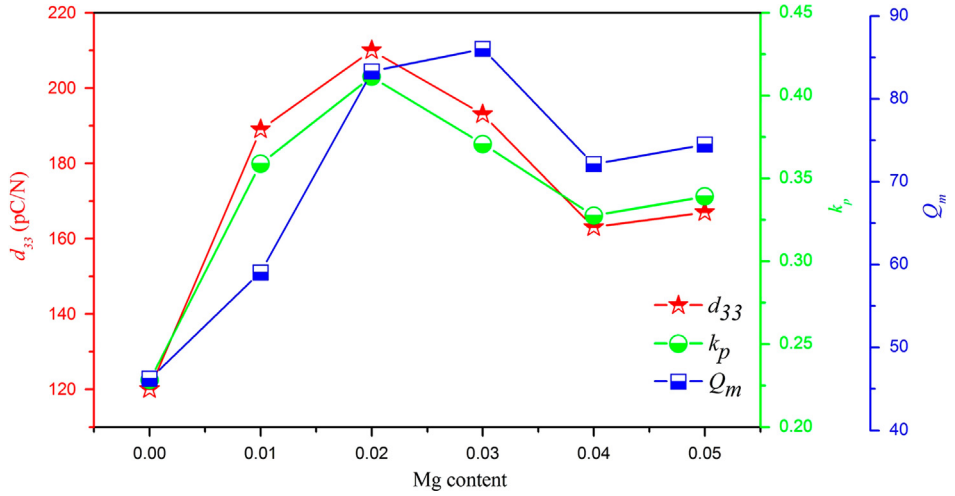

Fig. 8. Piezoelectric properties and mechanical quality factor of KNMN–BNT ceramics as a function of Mg content.

4. Conclusions

$0.975[(Na_{0.5}K_{0.5})_{1-2x}Mg_xNbO_3]$–$0.025(Bi_{0.5}Na_{0.5}TiO_3)$ (KNMN–BNT, $x=0$, 0.01, 0.02, 0.03, 0.04 and 0.05) lead-free piezoelectric ceramics were synthesized by the conventional solid-state reaction technique. The effects of the MgO addition on the crystal structure, dielectric, ferroelectric and piezoelectric properties were studied. XRD analysis revealed that the KNMN–BNT ceramics formed a single phase solid solution with perovskite structure with $x \leq 0.03$ and a MPB between orthorhombic and tetragonal phases could be observed with $0.02 \leq x \leq 0.05$. The T_{O-T} is less than 95 °C and the T_c is almost unchanged ($\sim$ 60 °C) as the MgO content increased. The ceramics with $x=0.02$ had enhanced ferroelectric and piezoelectric properties of $2E_c \sim 22.4$ kV/cm, $2P_r \sim 39.2$ μC/cm^2, $d_{33} \sim 210$ pC/N, and $k_p \sim 0.41$, together with the optimal dielectric properties of $\varepsilon_r \sim 637$ and tan $\delta \sim 0.09$, which may be due to the involvement of a MPB. The effect shows that the introduction of MgO is an effective method to improve the density as well as the electrical properties of KNMN–BNT ceramics, which may benefit the development of KNN-based ceramics.

Acknowledgment

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