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Ceramics International 40 (2014) 4389–4394

CERAMICS INTERNATIONAL

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Phase transition, microstructure, and dielectric properties of Li/Ta/Sb co-doped (K, Na)NbO_3 lead-free ceramics

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Received 5 July 2013; received in revised form 24 August 2013; accepted 26 August 2013
Available online 3 September 2013

Abstract

Li/Ta/Sb co-doped lead-free $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.93-x}Ta_xSb_{0.07})O_3$ (abbreviated $KNLNST_x$) piezoelectric ceramics, with Ta-doping ratio of x ranging from 0.0275 to 0.0675, were synthesized using the conventional solid-state reaction method at the sintering temperature of 1130 °C. The effects of Ta content on the microstructure, dielectric properties, and phase transition behavior of the prepared ceramics were systematically investigated. The X-ray diffraction results show that all $KNLNST_x$ ceramics formed a secondary phase, which is assigned to the tetragonal tungsten-bronze type (TTB) structure phase, and showed a phase transition from an orthorhombic symmetry to a tetragonal symmetry across a composition region of $0.0375 < x < 0.0475$. The grain shape and size that correspond to the phase structure transformations can be clearly observed in the scanning electron microscopy images. As x increased to 0.0475, the $KNLNST_{0.0475}$ ceramics changed from orthorhombic to tetragonal structure and showed excellent piezoelectric properties of $d_{33}=313$ pC/N, $k_p=47\%$, and $\varepsilon_r=1825$. By contrast, samples of $x=0.0375$ with orthorhombic symmetry exhibited poor piezoelectric properties, with $d_{33}=200$ pC/N and $\varepsilon_r=1015$. These results indicate that phase structure is vital in the piezoelectric properties of KNN lead-free ceramics.

Keywords: C. Dielectric properties; Lead-free ceramics; Microstructure; Phase transition

1. Introduction

(K, Na)NbO_3 (KNN) has been widely investigated as a potential alternative candidate for conventional lead-based piezoelectric materials with high Curie temperature and desirable piezoelectric properties [1,2]. However, obtaining dense and well-sintered pure KNN ceramics using an ordinary sintering process is difficult because of the high volatility of alkaline elements at high temperatures, which has restricted research progress [3,4]. Pure KNN ceramics, prepared by the conventional solid-state reaction method, show a poor d_{33} value of 80 pC/N [5]. Several methods have been devoted to the property and densification improvement of KNN ceramics, including the application of various fabrication techniques, such as hot-pressed sintering [6], spark-plasma sintering [7], microwave sintering [8], two-step sintering [9], sol–gel method [10], and cold-isostatic press [11]. Nevertheless, these techniques were found to be unsuitable for industrial use. Other techniques have employed a small amount of sintering materials, such as CuO [12], MnO_2 [13], $AgTaO_3$ [14], Fe_2O_3 [15], ZnO [16], and SnO_2 [17], to lower sintering temperature and enhance density. These techniques involve the presence of a liquid phase, but the piezoelectric properties of the ceramics are weakened.

Addition of Li/Ta/Sb can modify T_c, significantly improve the piezoelectric performance, and enhance stability and compactness of KNN ceramics. Excellent piezoelectric and electromechanical properties have been achieved in Li-modified KNN ceramics since Saeri et al. [18] reported KNN-based lead-free ceramics with a d_{33} value of 215 pC/N. Salto et al. [19] used the growth of reactive templates to investigate (K, Na)NbO_3–Li(Ta, Sb)O_3. The as-obtained ceramics exhibited piezoelectric properties as high as 416 pC/N, which is comparable with lead zirconate titanate ceramics. Partial substitutions of Li at the A-site and Sb and/or Ta at the B-site of perovskite ABO_3 structure can induce a sharp change in KNN lattice parameters, thereby facilitating phase transition. Achieving a crystal structure where both orthorhombic and tetragonal phases coexist would be advantageous for the improvement of the piezoelectric properties

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http://dx.doi.org/10.1016/j.ceramint.2013.08.110

of KNN-based ceramics. Jiang et al. [20] studied $(Na_{0.52}K_{0.48-x}Li_x)(Nb_{0.86}Ta_{0.10}Sb_{0.04})O_3$ ceramics with x varied from 0 to 0.08. As x shifted from 0.02 to 0.04, phase structure transformation from orthorhombic to tetragonal was observed. Furthermore, the piezoelectric properties were substantially elevated: d_{33} increased from ~195 pC/N to ~260 pC/N and ε_r increased from ~775 to ~1050. Yang et al. [21] observed the same phase-structure transition phenomenon by modifying the Ta content in $(K_{0.44}Na_{0.52}Li_{0.04})(Nb_{0.96-x}Ta_xSb_{0.04})O_3$. The $KNLNST_x$ ceramics with $x=0.2$ showed significantly enhanced dielectric properties of $d_{33}=252$ pC/N. However, Ta_2O_5 is more expensive than Sb_2O_3, thereby making it unsuitable for industrial production.

In the current study, $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.93-x}Sb_{0.07}Ta_x)O_3$, with relatively high Sb content and low Ta levels, was selected as the main research subject. We studied the effects of x variation from 0.0275 to 0.0675 on ceramic phase structure and piezoelectric properties.

2. Experimental procedure

K_2CO_3 (99%), Na_2CO_3 (99.8%), Li_2CO_3 (98%), Nb_2O_5 (99.5%), Ta_2O_5 (99.99%), and Sb_2O_3 (99.5%) were used as raw materials to prepare $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.93-x}Sb_{0.07}Ta_x)O_3$ ($x=0.0275$, 0.0375, 0.0475, 0.0575, 0.0675) ceramics using the conventional solid-state reaction method. The stoichiometric powders were mixed in ethanol by ball milling for 12 h and were subsequently dried and calcined at 900 °C for 5 h. The calcined powders were mixed with 3 wt% polyvinyl alcohol (PVA) solution and then uniaxially pressed into pellets with a diameter of 1.5 cm under 300 MPa. After burning the PVA, the green disks were sintered in air at selected temperatures for 3 h, depending on their x values. The microstructure was observed using scanning electron microscopy (SEM, JSM-5610LV/Noran-Vantage). Powder X-ray diffraction (XRD) (D8 Advance) was used to identify crystal structures and phases. For the measurement of dielectric and piezoelectric properties, silver paste electrodes were formed at the two circular surfaces of the disk-shaped specimens after firing at 700 °C for 10 min. The piezoelectric constant d_{33} was measured using a static piezoelectric constant testing meter (ZJ-3A, Institute of Acoustics, Chinese Academy of Science, Beijing, China). Dielectric properties, as a function of temperature and frequency, were measured using an impedance analyzer (HP4294A). Contrast between polarization and electric field hysteresis loops was measured using a ferroelectric tester (TF Analyzer 2000). The measurement of piezoelectric and electromechanical properties was conducted 24 h after the poling process.

3. Results and discussion

Fig. 1(a) shows the XRD patterns of the $KNLNST_x$ ceramics with x ranging from 0.0275 to 0.0675 sintered at 1130 °C for 3 h. Fig. 1(b) shows the magnified XRD patterns in the 2θ range of 44–48°. A secondary phase was detected in all doped samples when 2θ was approximately 28.5°. The secondary phase could be assigned to the tetragonal tungsten-bronze (TTB) type structure phase, which did not disappear as the composition changed. The occurrence of the TTB secondary phase was attributed to the volatilization and segregation of the alkali elements during the sintering process for Li/Ta-modified KNN material [22], which induced B-site ion excess that was accommodated through TTB phase formation. The combination of 2θ from 45° to 46° [Fig. 1(b)] evidently shows that ceramics with $x<0.0475$ possess an orthorhombic structure with the splitting of the (200)/(002) characteristic peaks. As x increased to 0.0475, a tetragonal phase appeared, and the tetragonal structure was maintained until $x=0.0675$. Fig. 2 reveals the lattice parameter evolution as a function of the Ta amount. The lattice parameters of a and b decreased, whereas that of c increased with increasing Ta^{5+} content, indicating tetragonal phase formation. This phenomenon occurs because Ta^{5+}, with a smaller radius (0.64 Å) and higher electronegativity, occupies Nb^{5+} (0.69 Å) sites.

Fig. 3 depicts the micrographs of the $KNLNST_x$ ceramics with $x=0.0275$, 0.0375, 0.0475, 0.0575, and 0.0675 sintered

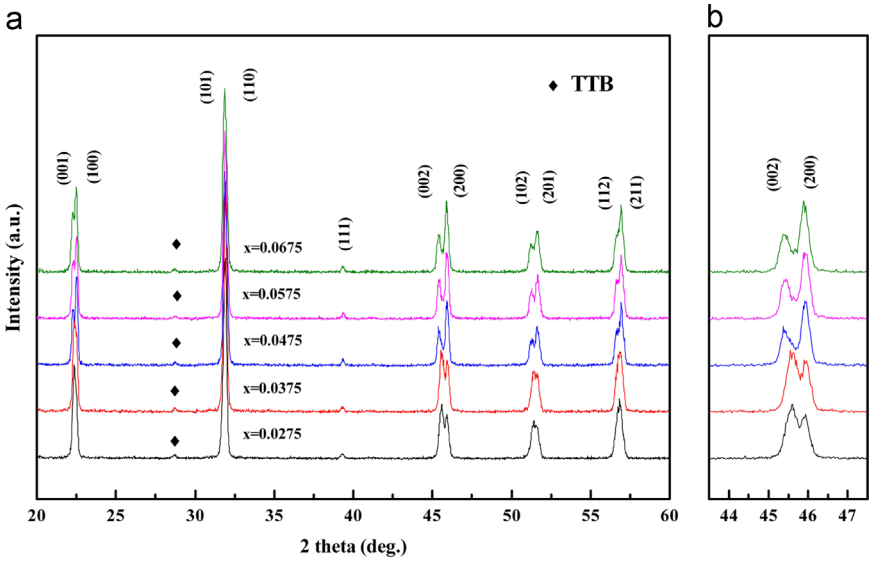

Fig. 1. XRD patterns of the $KNLNST_x$ ceramics with x from 0.0275 to 0.0675 in the range of 2θ: (a) from 20° to 60° and (b) from 44° to 48°.

at 1130 °C. Grains with similar spherical polyhedron shapes coexist with some rod-like grains (marked by arrows) that constitute the ceramics; the size of fine grains was approximately 1 μm and the microstructure remained relatively uniform, as shown in Fig. 3(a) and (b). Significant changes were observed in the morphology of the grains as x increased to 0.0475. Square grains gradually replaced the polyhedron grains and exhibited a bimodal distribution, with many fine grains located at the coarse grain boundaries [Fig. 3(c)–(e)]. Such abrupt changes in grain morphology was caused by the phase transformations detected by XRD for Ta content ranging from $x=0.0375$ to $x=0.0475$. Meanwhile, the amount of small grains with constant size (approximately 1 μm), increased with further increase in Ta content, which indicates that grain growth was inhibited and that average grain size decreased as Ta substituted Nb.

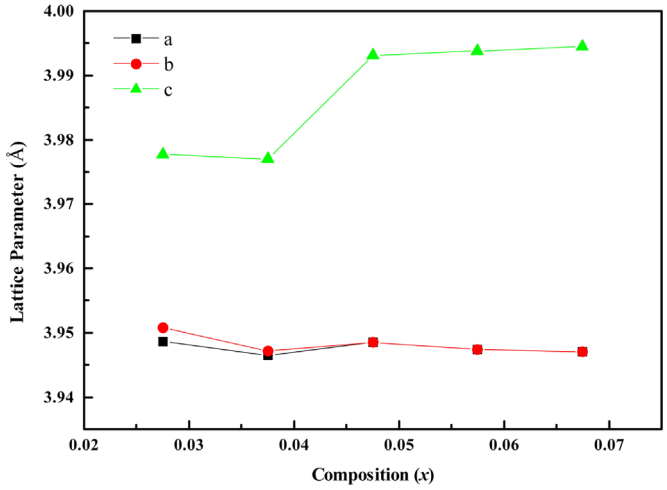

Fig. 2. Lattice parameter evolution as a function of the Ta amount.

Fig. 4 shows the dependence of measured densities as a function of Ta content. The measured densities of the samples initially increased with increasing Ta content and then decreased until $x=0.0475$. The maximum density of 4.58 g/cm^3 was obtained for the KNLNST$_x$ ceramic with $x=0.0375$. This density transition is attributed to the different theoretical densities of the different phases. Ceramics with tetragonal phase structures are more difficult to densify than those with orthorhombic phase structures, given that the grains of the former possess fewer crystal faces and more highlighted pointedness. With further increase in x, the measured densities of the samples increased again, which suggest that the amount of fine grains increases with Ta content. Likewise, the SEM images in Fig. 3(d) and (e) shows that these fine grains fill the gaps between large grains, thereby, enhancing ceramic density.

Fig. 5(a) shows the changes in d_{33} as a function of Ta^{5+} content. For KNLNST$_x$ ceramics, the d_{33} value was observed to increase rapidly when x changed from 0.0375 ($d_{33}\sim$200 pC/N) to 0.0475 ($d_{33}\sim$313 pC/N), whereas d_{33} decreased as x further increased to 0.0575. This variation trend is related to the MPB phase that exists in the composition of x between 0.0375 and 0.0475, according to the XRD results. The samples with MPB exhibit better piezoelectric coefficients d_{33} for the KNN-based ceramics. However, in the present study, the d_{33} value of $x=0.0475$ was noted to be significantly higher than the d_{33} value of $x=0.0375$. The values of $x=0.0575$ and $x=0.0675$, which deviate largely from the composition of the MPB, also achieved higher piezoelectric coefficients than that of $x=0.0375$. This result indicates that the bulks, which are near the tetragonal side of MPB, exhibit more excellent piezoelectric properties than the ones that are near the orthorhombic side of the MPB. Meanwhile, with increasing amount of Ta, the k_p of KNLNST$_x$ ceramics showed a relatively high and steady value of approximately 0.44–0.49 [Fig. 5(b)]; the slight increase in k_p

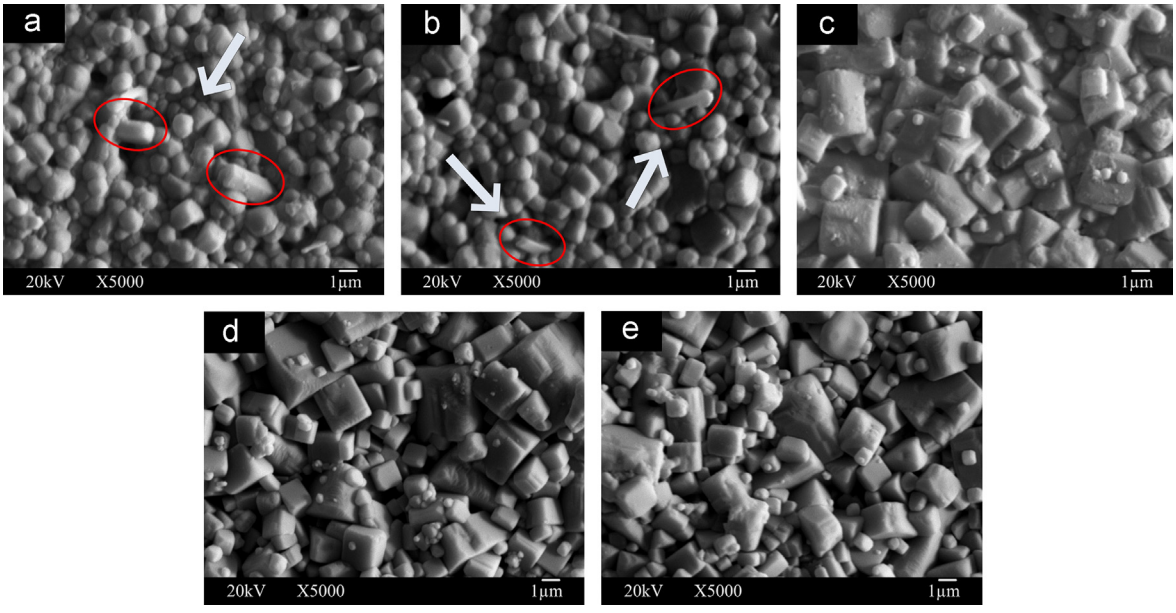

Fig. 3. SEM images of the KNLNST$_x$ ceramics sintered at 1130 °C with different Ta contents: (a) $x=0.0275$, (b) $x=0.0375$, (c) $x=0.0475$, (d) $x=0.0575$, and (e) $x=0.0675$.

can be seen when x was increased to 0.0375. Further increasing x to 0.0575 led to a decrease in k_p; however, a slight increase appeared at $x=0.0675$. Contrary to the change of the d_{33} value, the Q_m curve, which exhibits an obvious "valley" region, reached a minimum at $x=0.0475$ and then increased with further increasing Ta content [Fig. 5(c)]. Relative permittivity (ε_r) and dielectric loss ($tg\delta$) of the KNLNST$_x$ ceramics with different Ta contents are described in Fig. 5(d). ε_r and d_{33} were found to show an analogous variation tendency, resulting in a peak value at $x=0.0675$ (1867). $tg\delta$ decreased initially as x increased from $x=0.0275$ to $x=0.0475$ and fluctuated slightly until $x=0.0675$. Thus, the KNLNST$_x$ ceramics with $x=0.0475$ exhibited the optimum piezoelectric properties: $d_{33}\sim313$ pC/N, $\varepsilon_r\sim1825$, $k_p\sim0.47$, and $Q_m\sim29$.

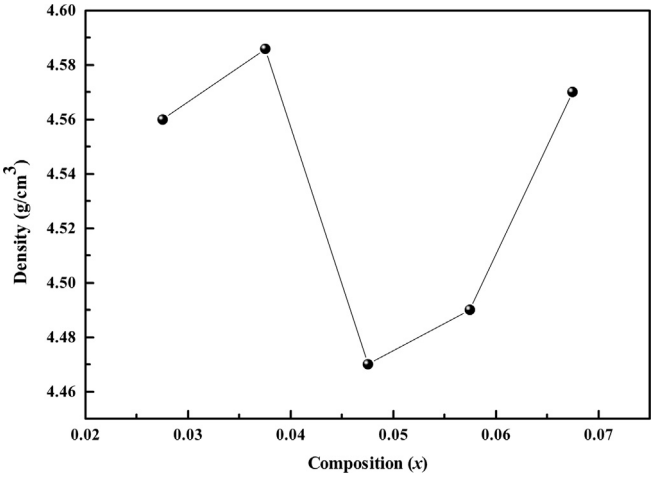

Fig. 4. Dependence of measured densities as a function of Ta content.

Fig. 6(a) and (b) shows the room polarization–electric field (P–E) hysteresis loops of the KNLNST$_x$ ceramics with two phase structures, (a) orthorhombic phase ($0.0275\le x\le0.0375$) and (b) tetragonal phase ($0.0475\le x\le0.0675$). With an electric field of 3 kV/mm, well-saturated P–E loops were attained for all the samples. The P–E loops of the bulks with orthorhombic phase were square-like, showing larger remanent polarization and smaller coercive field. As x increased to 0.0475, P_r started to decrease and E_c increased significantly; the P–E loops transformed into diamond shape. The optimum ferroelectric properties ($P_r=19.5$ μC/mm^2 and $E_c=913$ V/mm) were found at $x=0.0375$, wherein the d_{33} value was only 200 pC/N. This result is significantly lower than that of $x=0.0475$ ($d_{33}\sim313$ pC/N, $P_r=15.5$ μC/mm^2, and $E_c=1319$ V/mm). The KNN-based ceramics are generally known to possess high piezoelectric properties corresponding to higher remanent polarization and lower coercive field, which are contradictory to the results of this series of samples. Assuming that variation curves of the P_r and E_c values are divided into two parts [Fig. 6(c) and (d)], the samples of $x=0.0475$ are found to possess the highest P_r and the lowest E_c, corresponding to the highest value of d_{33} in the tetragonal phase. The same result is found for the samples of $x=0.0375$ in the orthorhombic phase. These observations probably suggest existing larger difference of P_r and E_c among

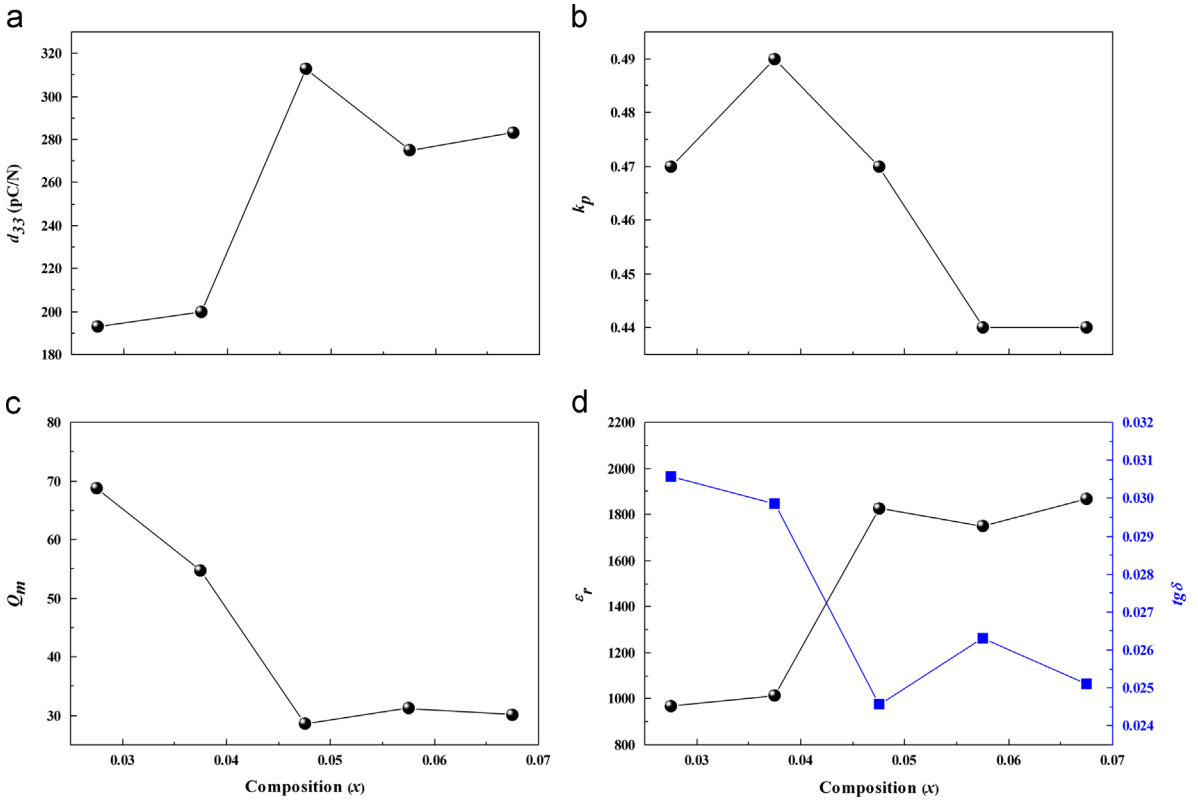

Fig. 5. Dependence of electrical properties of the KNLNST$_x$ ceramics with $x=0.0275$, 0.0375, 0.0475, 0.0575, and 0.0675 sintered at 1130 °C for 3 h: (a) piezoelectric coefficients (d_{33}), (b) planar mode electromechanical coupling coefficient (k_p), (c) mechanical quality factor (Q_m), (d) relative permittivity (ε_r) and dielectric loss ($tg\delta$).

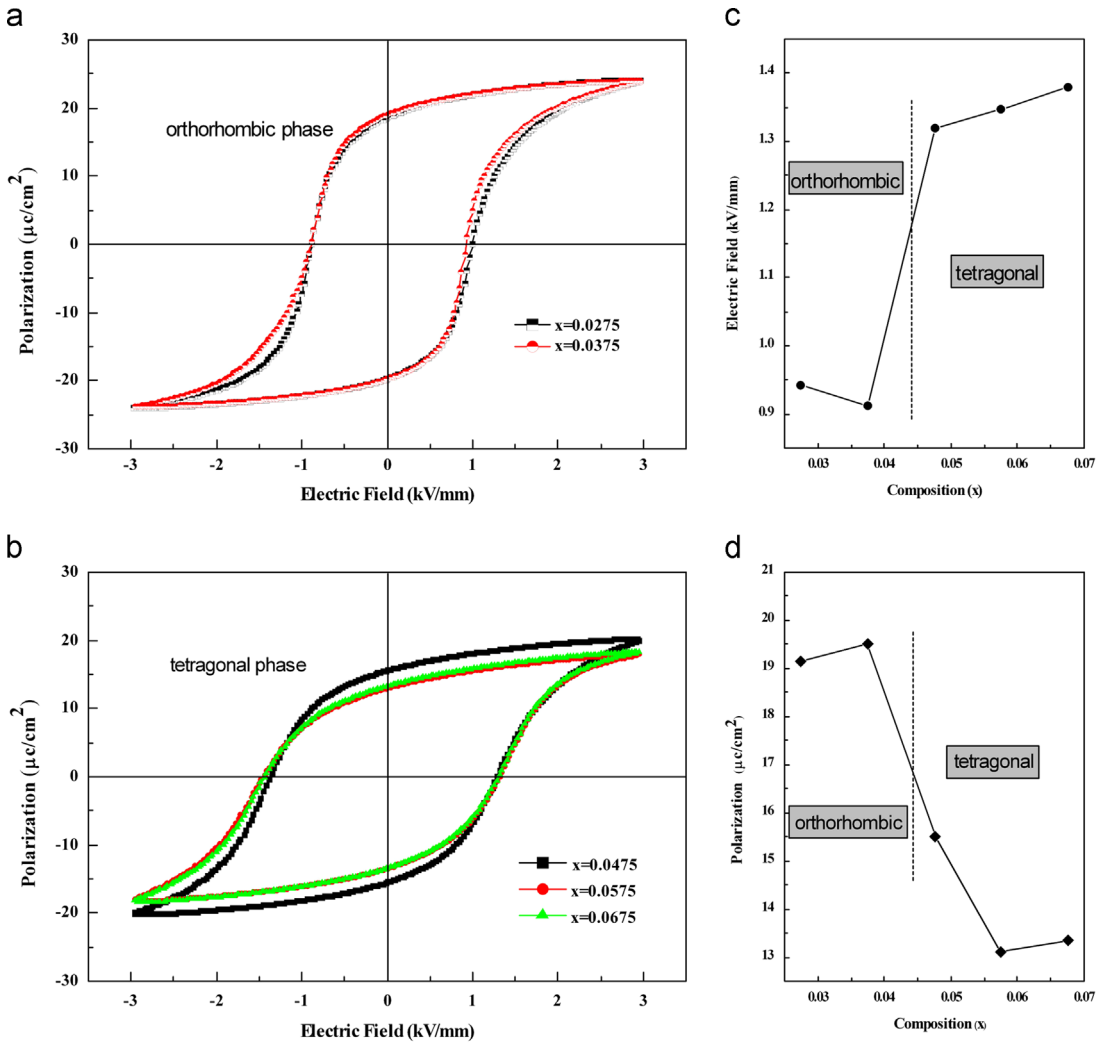

Fig. 6. (a) Polarization–electric field hysteresis loops of the $KNLNST_x$ ceramics with $x=0.0275$, 0.0375 sintered at 1130 °C for 3 h, (b) polarization–electric field hysteresis loops of the $KNLNST_x$ ceramics with x from 0.0475 to 0.0675 sintered at 1130 °C for 3 h, (c) E_c change of the $KNLNST_x$ samples as a function of Ta content, and (d) P_r values of the $KNLNST_x$ ceramics with x from 0.0275 to 0.0675.

different phase structures. Numerous studies consider that the significant increase in the coercive field of samples results from the formation of oxygen vacancies through the substitution of lower valence ions to Nb^{5+} sites. However, in the present study, the Ta and Sb valences were both equivalent to the Nb chemical valence. Therefore, the explanation of the related literature is not applicable to the present samples. Le et al. [23] reported that the coercive field is reduced significantly with decreasing domain size. In addition, obtaining finer domain sizes requires significantly smaller grain sizes [24]. As can be seen clearly in Fig. 3 (a)–(e), the amount of finer grains of the bulks with orthorhombic structure is higher than that of the ones with tetragonal phase structures. Thus, the variation in the coercive field E_c value could depend on grain size, which can be affected by phase structure variations. However, P_r value mutation among different phase structures is not well-understood and requires further study. P_r value mutation is also probably due to the domain size.

Fig. 7(a) shows the temperature dependence of dielectric constant (measured at 10 kHz) for the $KNLNST_x$ ceramics with $x=0.0475$. Two peaks can be detected in the temperature ranges of 50–100 °C and 250–300 °C, respectively, which corresponds to the transition temperatures of orthorhombic phase to tetragonal phase (T_{O-T}) and tetragonal phase to cubic phase (T_c). The T_{O-T} and T_c values for $x=0.0475$ are 83 °C and 275 °C, respectively. Fig. 7(b) shows that optimal d_{33} can be obtained when poling temperature is at 70 °C, which approaches T_{O-T}. This finding is attributed to the metastability of the phase structure near the T_{O-T}, where domains possess higher activity and are more likely to reverse. Moreover, d_{33} decreased monotonously from 313 pC/N to 268 pC/N as the polarization temperature continuously increased to 150 °C.

4. Conclusions

The effect of phase structure transformation caused by the Ta modification on the microstructure, dielectric, piezoelectric, and ferroelectric properties of the $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.93-x}Ta_xSb_{0.07})O_3$ ceramics was examined. Orthorhombic phases

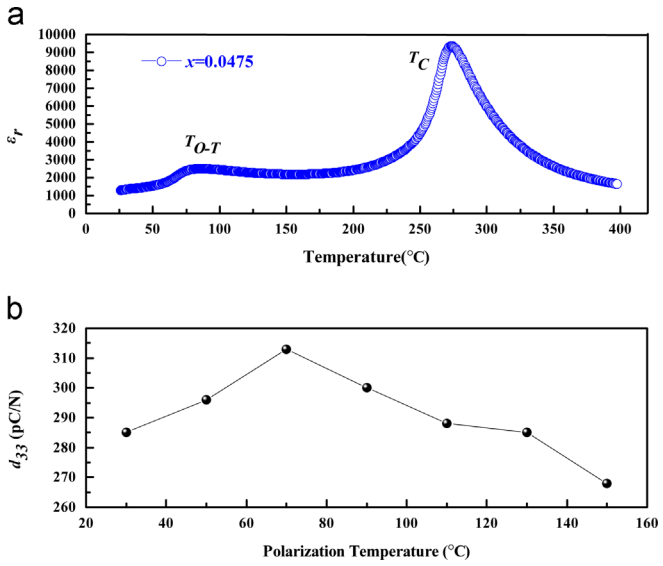

Fig. 7. (a) The temperature dependence of dielectric constant (measured at 10 kHz) for the $KNLNST_x$ ceramics with $x=0.0475$ and (b) poling temperature dependence.of the piezoelectric coefficients (d_{33}) for the $KNLNST_x$ ceramics with $x=0.0475$ sintered at 1130 °C for 3 h.

mainly exist in $KNLNST_x$ ceramics with $x=0.0375$ sintered at 1130 °C. In addition, apparent phase transition was observed as x increased to 0.0475, which was detected as the intensity of XRD peaks changed. The grain shape and size displayed a marked difference between orthorhombic phase and tetragonal phase. The bulks with tetragonal phase structures exhibited higher d_{33} than those with orthorhombic phase structures. Combining the effect of Ta content (4.75 mol%) and poling temperature (70 °C), the $KNLNST_x$ ceramics with $x=0.0475$ possess the optimum dielectric properties of $d_{33}=313$ pC/N, $k_p=0.47$, $\varepsilon_r=1824$, $P_r=15.5$ μC/mm^2, and $E_c=1319$ V/mm.

References

[1] W. Hu, X. Tan, K. Rajan, Combinatorial processing libraries for bulk $BiFeO_3$–$PbTiO_3$ piezoelectric ceramics, Applied Physics A 99 (2010) 427–431.

[2] C.M. Leung, S.W. Or, S.L. Ho, dc Magnetoelectric sensor based on direct coupling of Lorentz force effect in aluminum strip with transverse piezoelectric effect in $0.7Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.3PbTiO_3$ single-crystal plate, Journal of Applied Physics 107 (2010) (09E702 – 09E702 – 3).

[3] E. Cross, Lead-free at last, Nature 432 (2004) 24–25.

[4] R.E. Jaeger, L. Egerton, Hot pressing of potassium–sodium niobates, Journal of the American Ceramic Society 45 (1962) 209–213.

[5] L. Egerton, Dolores M. Dillon, Piezoelectric and dielectric properties of ceramics in the system potassium–sodium niobate, Journal of the American Ceramic Society 42 (1959) 438–442.

[6] K. Li, F.L. Li, Y. Wang, K.W. Kwok, H.L.W. Chan, Hot-pressed $K_{0.48}Na0.52Nb_{1-x}Bi_xO_3$ ($x=0.05$–0.15) lead-free ceramics for electro-optic applications, Materials Chemistry and Physics 131 (2011) 320–324.

[7] B.P. Zhang, J.F. Li, K. Wang, H.L. Zhang, Compositional dependence of piezoelectric properties in $Na_xK_{1-x}NbO_3$ lead-free ceramics prepared by spark plasma sintering, Journal of the American Ceramic Society 89 (2006) 1605–1609.

[8] Z.P. Xie, Z.L. Gui, L.T. Li, T. Su, Y. Huang, Microwave sintering of lead relaxor ferroelectric ceramics, Materials Letters 36 (1998) 191–194.

[9] X.M. Pang, J.H. Qiu, K.J. Zhu, J.Z. Du, (K, Na)NbO_3-based lead-free piezoelectric ceramics manufactured by two-step sintering, Ceramics International 38 (2012) 2521–2527.

[10] Y. Cao, K.J. Zhu, J.H. Qiu, H. Gu, Synthesis of potassium sodium niobate powders using an EDTA/citrate complexing sol–gel method, Particuology 10 (2012) 777–782.

[11] M. Ichiki, L. Zhang, M. Tanaka, R. Maeda, Electrical properties of piezoelectric sodium–potassium niobate, Journal of the European Ceramic Society 24 (2004) 1693–1697.

[12] B. Shao, J.H. Qiu, K.J. Zhu, H.H. Gu, H.L. Ji, Influence of sintering temperature on microstructure and electric properties of CuO doped alkaline niobate-based lead-free ceramics, Journal of Materials Science: Material in Electronics 23 (2012) 1455–1461.

[13] D.M. Lin, Q.J. Zheng, K.W. Kwok, C.G. Xu, C. Yang, Dielectric and piezoelectric properties of MnO_2-doped $K_{0.5}Na_{0.5}Nb_{0.92}Sb_{0.08}O_3$ lead-free ceramics, Journal of Materials Science: Materials in Electronics 21 (2010) 649–655.

[14] Y.Y. Wang, Q.B. Liu, F. Zhao, Phase transition behavior and electrical properties of $[(K_{0.50}Na_{0.50})_{1-x}Ag_x](Nb_{1-x}Ta_x)O_3$ lead-free ceramics, Journal of Alloys and Compounds 489 (2010) 175–178.

[15] W.L. Yang, Z.X. Zhou, B. Yang, R. Zhang, Z. Wang, H.Z. Chen, Y.Y. Jiang, Structure and piezoelectric properties of Fe-doped potassium sodium niobate tantalate lead-free ceramics, Journal of the American Ceramic Society 94 (2011) 2489–2493.

[16] F. Rubio-Marcos, J.J. Romero, M.G. Navarro-Rojero, J.F. Fernandez, Effect of ZnO on the structure, microstructure and electrical properties of KNN-modified piezoceramics, Journal of the European Ceramic Society 29 (2009) 3045–3052.

[17] Z. Li, G. Xu, Y. Li, A. Sun, L. Duan, J. Jiang, P. Cui, Dielectric and piezoelectric properties of ZnO and SnO_2 co-doping $K_{0.5}Na_{0.5}NbO_3$ ceramics, Physica B: Condensed Matter 405 (2010) 296–299.

[18] M.R. Saeri, A. Barzegar, H. Ahmadi Moghadam, Investigation of nano particle additives on lithium doped KNN lead free piezoelectric ceramics, Ceramics International 37 (2011) 3083–3087.

[19] Y. Saito, H. Takao, T. Tani, T. Nonoyama, K. Takatori, T. Homma, T. Nagaya, M. Nakamura, Lead-free piezoceramics, Nature 432 (2004) 84–87.

[20] X.P. Jiang, X.P. Hu, F.L. Jiang, X.D. Liu, R.Q. Yin, Li-modified sodium potassium tantalum niobate lead-free piezoelectric ceramics, Journal of Inorganic Materials 3 (2007) 465–468.

[21] Z.P. Yang, Y.F. Chang, L.L. Wei, Phase transitional behavior and electrical properties of lead-free $(K_{0.44}Na_{0.52}Li_{0.04})(Nb_{0.96-x}Ta_xSb_{0.04})O_3$ piezoelectric ceramics, Applied Physics Letters 90 (2007) 042911.

[22] Y. Wang, D. Damjanovic, N. Klein, N. Setter, High-temperature instability of Li and Ta modified (K,Na)NbO_3 piezoceramics, Journal of the American Ceramic Society 91 (2008) 1962–1970.

[23] D.T. Le, S.J. Kwon, N.R. Yeom, Y.J. Lee, Y.H. Jeong, M.P. Chun, J.H. Nam, J.H. Paik, B.I. Kim, J.H. Cho, Effects of the domain size on local d_{33} in tetragonal $(Na_{0.53}K_{0.45}Li_{0.02})(Nb_{0.8}Ta_{0.2})O_3$ ceramics, Journal of the American Ceramic Society 96 (2013) 174–178.

[24] W.W. Cao, C.A. Randall, Grain size and domain size relation in bulk ceramic ferroelectric materials, Journal of Physics and Chemistry of Solids 57 (1996) 1499–1505.