Structure and Properties of V_2O_5-doping KNN-LS-BF Piezoelectric Ceramics Sintered for Various Time

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Keywords: Piezoelectric ceramics, V_2O_5-doping, KNN-LS-BF, Sintering time

Abstract. 0.6mol% V_2O_5-doping $0.996(0.95Na_{0.5}K_{0.5}NbO_3\text{-}0.05LiSbO_3)\text{-}0.004FeBiO_3$ ((KNN-LS-BF)-V_2O_5) piezoelectric ceramics were synthesized by the solid-state reaction process, and the influence of sintering time on structure and properties of (KNN-LS-BF)-V_2O_5 ceramics were studied. It was found that all the samples sintered for various time are perovskite structure mixed with orthorhombic symmetry phase and tetragonal phase, but the sintering time has significant influence on the crystalline and properties. When the sintering time increases from 2 hours to 6 hours, the grain of (KNN-LS-BF)-V_2O_5 ceramics becomes more homogeneous and more tight-arrangement. The results revealed that the longer sintering time than 4 hours is beneficial for improving partial properties, such as d_{33}, $tan\delta$ and Q_m, but is adverse to ε_r and k_p, the (KNN-LS-BF)-V_2O_5 ceramics with optimum properties can be synthesized for 6 hours at 1080°C.

Introduction

Piezoelectric materials have been widely used for piezoelectric actuators, sensors and transducers, in which the lead oxide-based piezoelectric ceramics, such as lead zirconate titanate (PZT), have been widely investigated [1]. However, the use of lead in materials has been restricted due to its serious environmental pollution by law in more and more countries [2,3]. Therefore, the development of lead-free piezoelectric materials has attracted much attention and many research programs for them have been carried out.

Recent years, $K_{0.5}Na_{0.5}NbO_3$ (KNN) as one of the possible substitution of lead-based piezoelectric ceramics has increasingly attracted the attention of researchers [3,4]. However, it was found that the evaporation of Na and K in KNN-based ceramics sintered at high temperatures would degrade their piezoelectric properties [5,6]. Recent research has shown that additives of V_2O_5 can help improve the sintering behavior of $0.996(0.95Na_{0.5}K_{0.5}NbO_3\text{-}0.05LiSbO_3)\text{-}0.004BiFeO_3$ (KNN-LS-BF) piezoelectric ceramics, enhance the piezoelectric and dielectric properties [7].

In this paper, 0.6mol% V_2O_5-doping KNN-LS-BF ceramics were synthesized by the solid-state sintering ceramics technique, and the effects of sintering time on their structure and properties were investigated.

Experiments

The 0.6mol% V_2O_5-doping $[0.996(0.95Na_{0.5}K_{0.5}NbO_3\text{-}0.05LiSbO_3)\text{-}0.004FeBiO_3]$ ((KNN-LS-BF)-V_2O_5) ceramics were synthesized by solid-state reaction technique. According to stoichiometric molecular formula, the raw materials, including Na_2CO_3, K_2CO_3, Nb_2O_5, Li_2CO_3, Sb_2O_3, Fe_2O_3, Bi_2O_3 and V_2O_5, were weighed and then mixed them with ZrO_2 balls for 12 hours, using ethanol as the medium. The mixed powders were dried and calcined at 880°C for 6 hours. Using 5*wt*% PVA as binder, the mixed powders had been milled again for 24 hours and then dried again. The obtained powders were pressed into disks with 18 mm in diameter and 1.5 mm in thickness under 100MPa. The disks sintered for 2-6 hours at 1080°C, respectively. Silver paste as electrodes was applied on the

top and bottom surfaces of the ceramics. The polarization was carried out in the silicone oil with a temperature of 80°C at 3kV/mm for 20 minutes.

The crystal structure of samples was probed by X-ray diffraction (XRD) with Cu-Kα radiation (AXS D8-ADVANCE, Bruker). The surface microstructures and grain size were observed by a scanning electron microscope (SEM, JSM-5610LV, JEOL). The piezoelectric constant d_{33} was measured by a quasi-static piezoelectric d_{33} meter (ZJ-3A, CAS, Shanghai, China). The dielectric constant ε_r and the dielectric loss $tan\delta$ were obtained by measuring the capacitance and loss using the impedance analyzer (4294A, Agilent Inc., Bayan, Malaysia). The planar coupling coefficient k_p and the mechanical quality factor Q_m were determined by the resonance-anti-resonance technique using the impedance analyzer.

Results and Discussion

Fig. 1 is the XRD patterns of (KNN-LS-BF)-V_2O_5 ceramics sintered at 1080°C for different time from 2 hours to 6 hours. It can be seen that all the samples sintered for various time are perovskite structure mixed with orthorhombic symmetry phase and tetragonal phase, and some other phases, such as KVO_3 and $NaNbO_3$, are also observed, which indicated that the phase structure of (KNN-LS-BF)-V_2O_5 ceramics is different from the orthorhombic symmetry as reported for the pure KNN and the sintering time has insignificant influence on the phase structure of (KNN-LS-BF) -V_2O_5). The phase structure has not changed with the increase of sintering time, but the intensity of characteristic diffraction peaks in (KNN-LS-BF)-V_2O_5 ceramics become higher with the increase of sintering time from 2 hours to 6 hours, which means that the crystalline become better with the increase of sintering time. In addition, from Fig.1(b), it can be seen that the peaks in XRD patterns of (KNN-LS-BF)-V_2O_5 shifted from lower angles to higher angles first when the sintering time is below 4 hours, then shifted from higher angles to lower angles with the further increasing sintering time, which means a change in the crystal lattice constant of specimens, being the result of the change in diffusion level of V^{5+} ion into the KNN lattice.

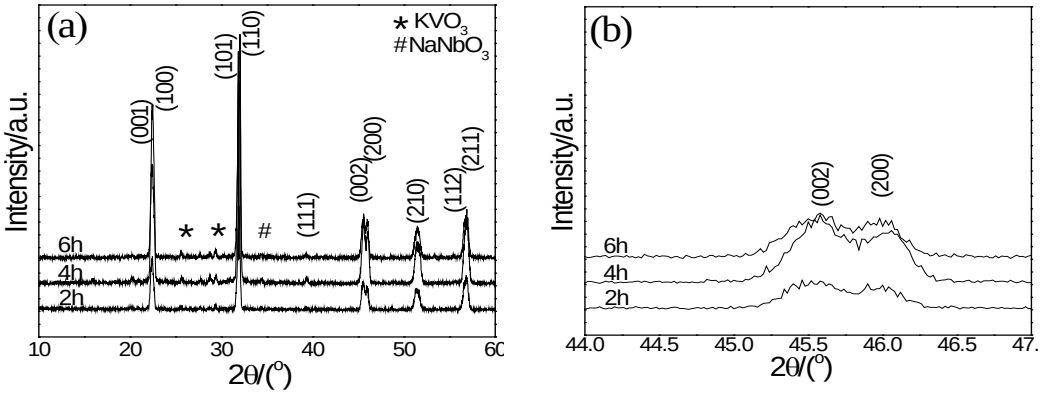

Fig.1 XRD patterns of (KNN-LS-BF)-V_2O_5 ceramics sintered for different time

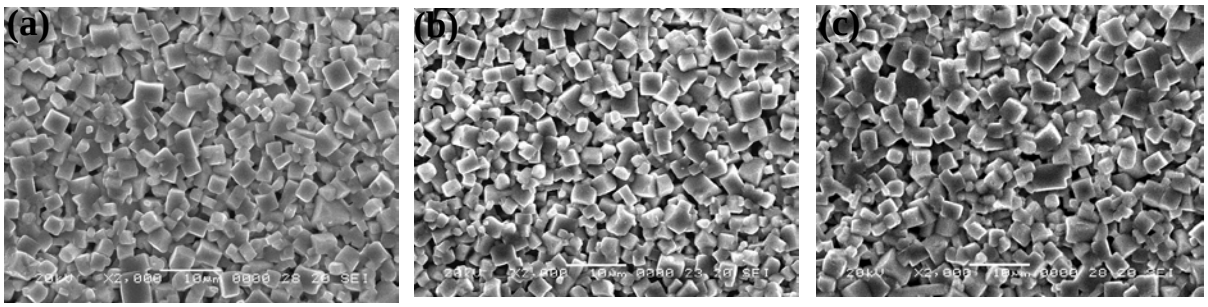

Fig.2 SEM images of (KNN-LS-BF)-V_2O_5 ceramics sintered for: (a) 2 hours (b) 4 hours (c) 6 hours

Fig.2 is the SEM images of (KNN-LS-BF)-V_2O_5 ceramics sintered at 1080°C for different time. It can be seen that the grain size is similar which indicated that the influence of sintering time on the grain size is not obvious. When the increasing of sintering time from 2 hours to 6 hours, the grain of (KNN-LS-BF)-V_2O_5 ceramics becomes more homogeneous and more tight-arrangement. However, when the sintering time is below 4 hours, there are some holes in the grain boundary, meaning degrade in density, which indicate that insufficient sintering time is not effective in improving the density of (KNN-LS-BF)-V_2O_5 ceramics.

Fig.3 is the curves of piezoelectric constant d_{33} and planar electromechanical coupling coefficient k_p of (KNN-LS-BF)-V_2O_5 ceramics sintered at 1080°C dependent on the sintering temperature. With increasing sintering time, it is found that the d_{33} retain a stable value of 120 when the sintering time is below 4 hours, and then increase rapidly to the maximum values of 176 with the further increase of sintering time. Meanwhile, the k_p increase to a maximum values of 0.28 with the increase of sintering time from 2 hours to 4 hours, then decrease with the further increase of sintering time. The enhanced piezoelectric properties in the specimen sintered for 6 hours should be ascribed to the dense structure which has been confirmed in Fig.2.

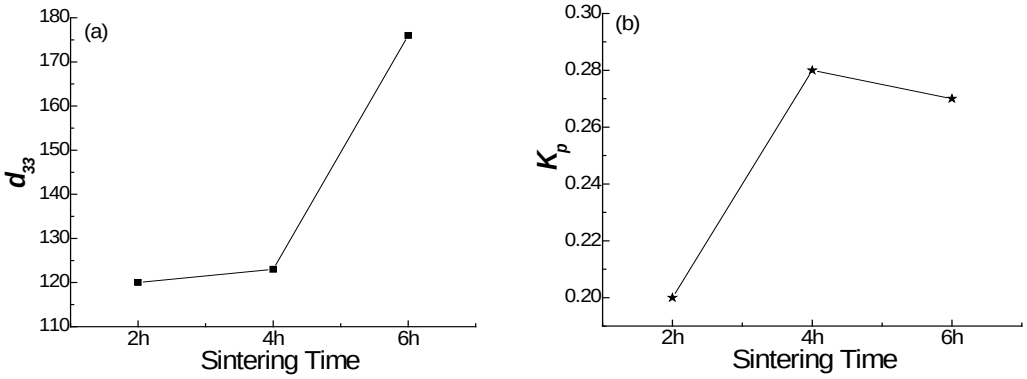

Fig.3 d_{33} and k_p of (KNN-LS-BF)-V_2O_5 ceramics sintered for different hours

Fig.4 is the values of dielectric constant ε_r and dielectric loss $tan\delta$ of (KNN-LS-BF)-V_2O_5 ceramics sintered at 1080°C for 2-6 hours. When the sintering time increase from 2 hours to 4 hours, it can be seen, like the variation trend of k_p value, that the ε_r and the $tan\delta$ both increases rapidly to a value of 491 and 12%, respectively, then decrease with the further increase of sintering time.

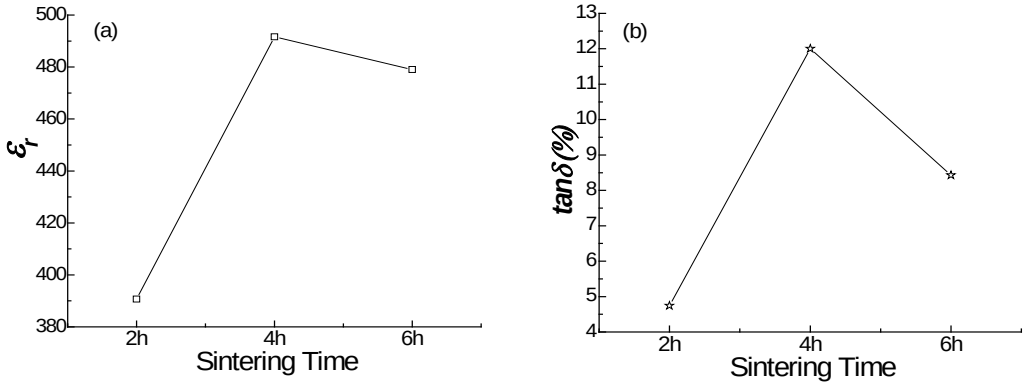

Fig.4 ε_r and $tan\delta$ of (KNN-LS-BF)-V_2O_5 ceramics sintered for different hours

Fig.5 shows the values of mechanical quality factor Q_m of (KNN-LS-BF)-V_2O_5 ceramics sintered at 1080°C for 2-6 hours. Being contrary to the ε_r, it can be seen that the value of Q_m decreases with the increase of sintering time from 2 hours to 4 hours, then increase with the further increase of sintering time. The maximum value of 78 obtained when the sintering time is 6 hours. These results indicate that the longer sintering time more than 4 hours is beneficial for improving partial properties, such as

the piezoelectric constant d_{33}, the dielectric loss $tan\delta$ and the mechanical quality factor Q_m, but it is adverse for improving the dielectric constant ε_r and the planar electromechanical coupling coefficient k_p. In general, the (KNN-LS-BF)-V_2O_5 ceramics with optimum combination properties can be synthesized for 6 hours at 1080°C.

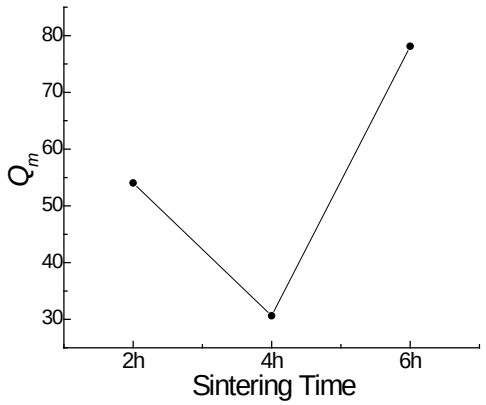

Fig.5 Q_m of (KNN-LS-BF)-V_2O_5 ceramics sintered for different hours

Summary

All (KNN-LS-BF)-V_2O_5 ceramics sintered for various time from 2 hours to 6 hours are perovskite structure mixed with orthorhombic symmetry phase and tetragonal phase. With increase of the sintering time from 2 hours to 6 hours, the grain of (KNN-LS-BF)-V_2O_5 ceramics becomes more homogeneous and more tight-arrangement. The results revealed that the longer sintering time more than 4 hours is beneficial for improving partial properties, such as d_{33}, $tan\delta$, Q_m, but is adverse to ε_r and k_p. The optimum properties can be obtained in the specimen sintered for 6 hours at 1080°C.

Acknowledgements

This work was financially supported by the Guangxi Nature Science Foundation (2010GXNSFD013007).

References

[1] D.W.Wang, M.S.Cao, S.J.Zhang, Investigation of ternary system Pb(Sn,Ti)O_3-Pb($Mg_{1/3}Nb_{2/3}$)O_3 with morphotropic phase boundary compositions, J. Eur. Ceram. Soc. 32 (2012) 441-448.

[2] Y. Lin, L. Zhang, J. Yu, Stable piezoelectric property of modified $BiFeO_3$-$BaTiO_3$ lead-free piezoceramics, J. Mater. Sci.: Mater. Electron. 26 (2015) 8432-8441.

[3] R.Z. Zuo, C. Ye, X.S. Fang, $Na_{0.5}K_{0.5}NbO_3$-$BiFeO_3$ lead-free piezoelectric ceramics, J. Phys. Chem. Solids 69 (2008) 230-235.

[4] J.M.Jin, D.D.Wan, Y.Yang, Q.Lin, M.Zha, A linear ultrasonic motor using $(K_{0.5}Na_{0.5})NbO_3$ based lead-free piezoelectric ceramics, Sensor. Actuat. A, 165(2011)410-414.

[5] S Wongsaenmai, K Kanchiang, S Chandarak, Crystal structure and ferroelectric properties of Mn-doped $((K_{0.5}Na_{0.5})_{0.935}Li_{0.065})NbO_3$ lead-free ceramics, Curr. Appl. Phys.12 (2012) 418-421.

[6] H.Q.Wang, D.S.Ruan, Y.J.Dai, X.W.Zhang, Relationship between phase structure and electrical properties of $(K_{0.5}Na_{0.5})NbO_3$-$LiTaO_3$ lead-free ceramics, Curr. Appl.Phys. 12 (2012) 504-508.

[7] H.Wang, X.Zhai, J.Xu, C.Yuan, L.Yang, Temperature stability of V_2O_5-doped KNN-LS-BF lead-free piezoelectric ceramics, J. Mater. Sci.: Mater. Electron. 26 (2015) 2556-2559.

