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Letter

Lead-free (K, Na)NbO_3–$Bi_{0.5}K_{0.5}ZrO_3$–$BaZrO_3$ ternary system: Microstructure and electrical properties

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ABSTRACT

In this work, new (K, Na)NbO_3 ternary system {e.g., $(1-x)$ $(0.96K_{0.48}Na_{0.52(1+3/995)}NbO_3$–$0.04Bi_{0.5}K_{0.5}ZrO_3)$–$x$$BaZrO_3$} lead-free piezoceramics have been prepared by the conventional solid state sintering method, and effects of $BaZrO_3$ contents on their microstructure and electrical properties were investigated in detail. The rhombohedral–orthorhombic and orthorhombic–tetragonal phase coexistence has been found in the ceramics with the composition range of $0.0125 < x \leqslant 0.02$, and then the ceramics with $x = 0.0175$ exhibit enhanced electrical properties (e.g., $d_{33} = 305$ pC/N, $k_p = 49.5\%$, $\varepsilon_r = 1945$, $\tan\delta = 3.9\%$, $P_r = 22.5$ μC/cm^2, and $E_C = 10.2$ kV/cm). In addition, such a ceramic possesses a high Curie temperature ($T_C = 300$ °C). These results show that this material should be one of the most promising lead-free candidates for piezoelectric applications.

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1. Introduction

For protecting environment, $K_{0.5}Na_{0.5}NbO_3$ (KNN) lead-free piezoceramics have been considered as one of the most promising candidates for replacing the lead-based ones, owing to its high Curie temperature and enhanced electrical properties [1,2]. However, a pure KNN ceramic, which was prepared by the conventional solid-state reaction method, shows a poor d_{33} value of 80 pC/N [1]. Therefore, it is highly expected to develop high performance KNN-based lead-free ceramics. It is well known that the excellent piezoelectric properties of Pb(Zr, Ti)O_3 (PZT) are usually attributed to the formation of phase boundaries. As a result, it is of great interest to ask if it is possible to attain a high piezoelectricity in KNN by constructing new phase boundary. According to the research development of the construction of phase boundaries (e.g., R–O and O–T) [2–7], it seems reasonable to create a phase boundary consisting of R–O and O–T near room temperature.

In this study, we choose both $Bi_{0.5}K_{0.5}ZrO_3$ and $BaZrO_3$ to modify KNN ceramics to design new ternary material systems of $(1-x)(0.96K_{0.48}Na_{0.52(1+3/995)}NbO_3$–$0.04Bi_{0.5}K_{0.5}ZrO_3)$–$x$$BaZrO_3$ {$(1-x)$(0.96KNN–0.04BKZ)–xBZ}, and new phase boundaries (e.g., R–O and O–T) near room temperature were also obtained. In which, $Bi_{0.5}K_{0.5}ZrO_3$ is selected to decrease T_{O-T} to be near room temperature [8,9], and the $BaZrO_3$ is used to increase T_{R-O} to be around room temperature [4]. We hope to get the R–O and O–T phase coexistence by controlling the compositions of BKZ and BZ, and then a large d_{33} of 305 pC/N was observed. In addition, the relationship between phase boundary and electrical properties was discussed, and the underlying physical mechanisms were also given.

2. Experimental procedure

$(1-x)$(0.96KNN–0.04BKZ)–xBZ (x = 0, 0.005, 0.01, 0.0125, 0.015, 0.0175, and 0.02) ceramics were prepared by the conventional solid-state method, and Na_2CO_3 (99.8%), K_2CO_3 (99%), Nb_2O_5 (99.5%), Bi_2O_3 (99%), $BaCO_3$ (99.0%), and ZrO_2 (99%) were used as starting raw materials. The powders were mixed and milled with zirconia balls for 24 h, then dried and calcined at 850 °C for 6 h. The dried mixtures were added with 8 wt% PVA as a binder for granulation and then pressed into disks of 10 mm in diameter and 1.0 mm in thickness under 10 Mpa. After burning off PVA, these disks were sintered in the temperature range of 1100–1130 °C for 3 h in air. Silver paste electrodes were coated on both sides of these sintered samples and fired at 700 °C for 10 min. These disks were poled at room temperature in a silicone oil bath under a *dc* electric field of 3–4 kV/mm for10 min.

The phase structure of the specimens was determined by X-ray diffraction (DX-2000, Dandong, PR China), and their grain morphology was characterized by the field emission-scanning electron microscopy (FE-SEM) (JSM-7500, Japan). Dielectric behaviors were measured by an *LCR* meter (HP 4980, Agilent, USA), their piezoelectric constant d_{33} was obtained by a piezo-d_{33} meter (ZJ-3A, China), and the electromechanical coupling factor k_p were determined by an impedance analyzer (HP 4294A). The *P*–*E* hysteresis loops were tested at 10 Hz by a Radiant Precision Workstation (USA).

3. Results and discussion

Fig. 1(a) shows the XRD patterns of the ceramics with different BZ content, measured at room temperature and $2\theta = 20$–$60°$. All

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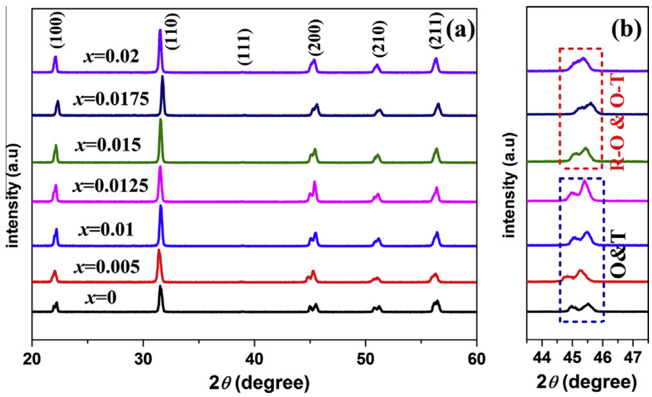

Fig. 1. (a) XRD patterns and (b) amplified XRD patterns of the ceramics.

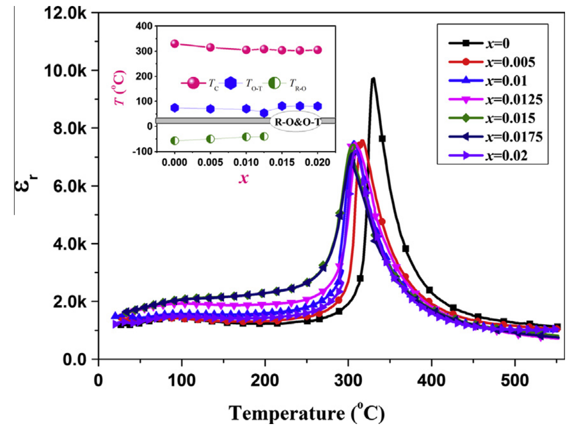

Fig. 3. (a) Temperature dependence of the ε_r of the ceramics, measured at10 kHz and room temperature ∼550 °C, and the insert is phase diagram.

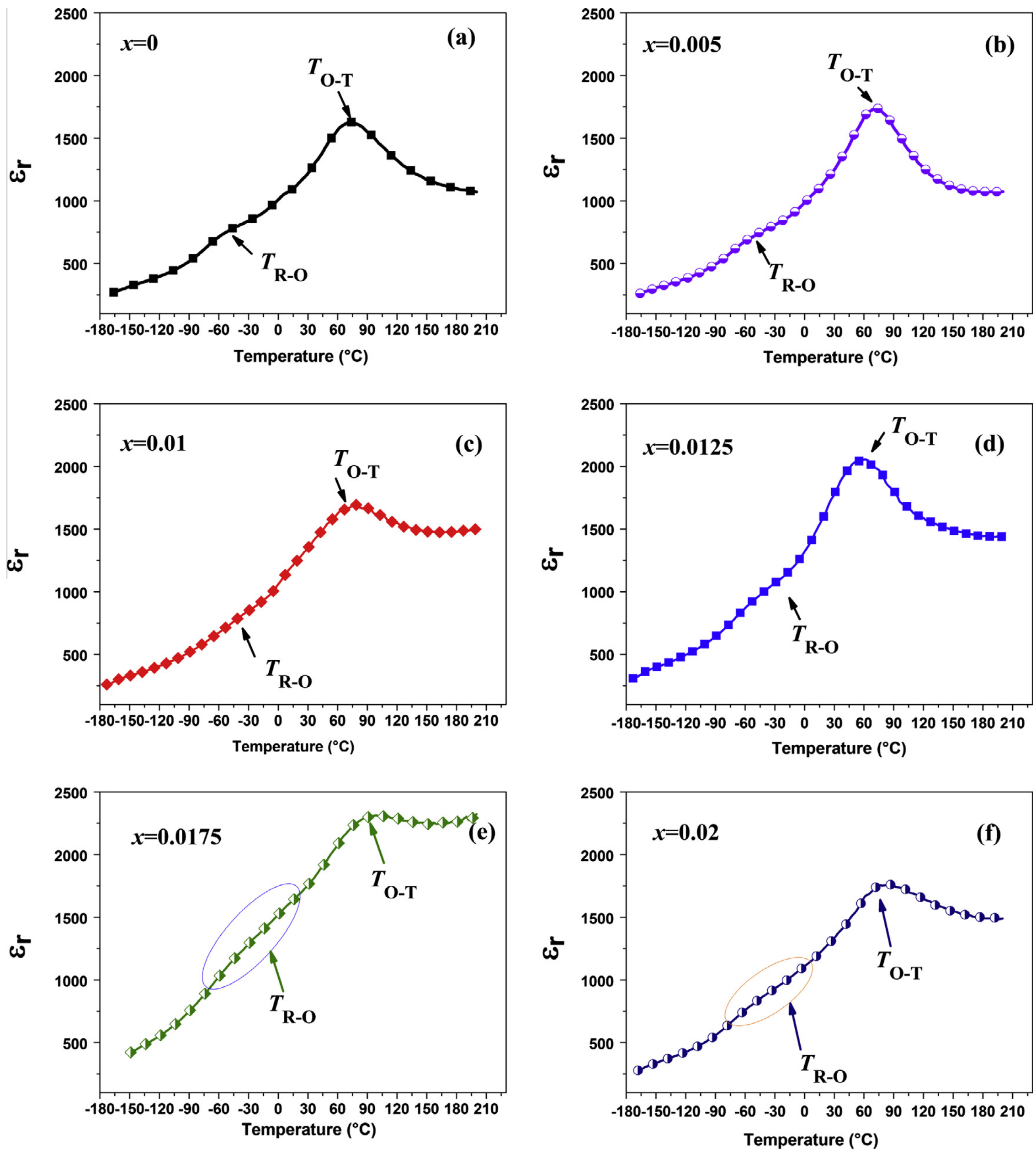

Fig. 2. Temperature-dependent dielectric constant of the ceramics in the temperature from −150 °C to 200 °C: (a) $x = 0$, (b) $x = 0.005$, (c) $x = 0.01$, (d) $x = 0.0125$, (e) $x = 0.0175$ and (f) $x = 0.02$.

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