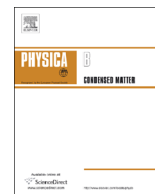




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Piezoelectric properties and diffusion phase transition around PPT of La-doped $(\text{Na}_{0.52}\text{K}_{0.44}\text{Li}_{0.04})\text{Nb}_{0.8}\text{Ta}_{0.2}\text{O}_3$ lead-free piezoelectric ceramics



Wenlong Yang*, Li Wang, Haidong Li, Junsheng Han, Hanjiang Xiu, Zhongxiang Zhou

Department of Applied Science, Harbin University of Science and Technology, Harbin 150080, PR China

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ABSTRACT

Lead-free ceramics $(\text{Na}_{0.52}\text{K}_{0.44}\text{Li}_{0.04})_{1-3x}\text{La}_x\text{Nb}_{0.8}\text{Ta}_{0.2}\text{O}_3$ (KNLNT-Lax, $x=0.00, 0.25, 0.5, 0.75, 1.00, 1.25$ mol%) as non-polluting materials were prepared by solid state reaction method. The structure, piezoelectric properties and temperature stability of KNLNT ceramic with different La doping concentrations were investigated. The results show a transition from orthorhombic-tetragonal mix phase to tetragonal single phase with the variation of La^{3+} concentrations. The SEM micrographs of surface and fractured surface show a dense microstructure with few micropores. The La-doped KNLTN ceramic will be an alternative candidate contributes to excellent piezoelectric properties, which are found in the 0.75 mol% La-doped KNLNT ceramics, with $d_{33}=215\text{pC/N}$, $k_p=42.8\%$ and $Q_m=89$. It has been remarkably improved that the temperature stability of KNLTN-Lax piezoelectric properties at room temperature, and the dielectric relaxation can be observed obviously. The mechanism of La doping was analyzed in terms of valence compensation and polymorphic phase transition (PPT) diffusion. The orthorhombic-tetragonal phase transition around room temperature and the relaxation transition were considered contributing to the excellent piezoelectric performance and improved temperature stability of La^{3+} -doped KNLTN.

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1. Introduction

Lead-free piezoelectric materials have been widely investigated in the past decade because of their excellent piezoelectric and dielectric properties. The (K, Na) NbO_3 (KNN) crystal and ceramics with high Curie temperature at about 420 °C were considered as potential alternative materials for the lead-based ones, which possesses orthorhombic phase at room temperature, and orthorhombic-tetragonal phase transition around 220 °C [1]. The (K, Na) NbO_3 (KNN)-based piezoelectric ceramics with Li, Ta, and Sb doped have drawn much attention in recent years [2–5]. The orthorhombic-tetragonal phase transition temperature of KNN ceramics could be moved from 220 °C down to room temperature through Li, Ta, and Sb doping, then more conditions of spontaneous polarization could be achieved due to phase coexistence, which will results in the excellent piezoelectric properties around polymorphic phase transition [6]. Although, further studies show that the PPT in KNN-based piezoelectric ceramics is different from the morphotropic phase boundary (MPB), it is still efficient to improve the piezoelectric properties of the material [7]. Y. Saito

et al. have obtained the structure of $(\text{K}_{0.44}\text{Na}_{0.52}\text{Li}_{0.04})(\text{Nb}_{0.8}\text{Ta}_{0.1}\text{Sb}_{0.04})\text{O}_3$ ceramic with d_{33} of more than 400 pC/N [2]. The piezoelectric properties of the complicated structure of (K, Na, Li) (Nb, Ta, Sb) O_3 ceramic would be improved on account of the existence of PPT at room temperature while the temperature stability would be reduced. However, the piezoelectric ceramics will inevitably encounter problems of temperature dependence in application process, so it is indispensable to propose solutions for temperature stability. It have been a significant investigation focus on both keeping high piezoelectric responses and excellent temperature stability simultaneously through PPT and/or MPB controlling and additives doping in the complicated structure of (K, Na, Li)(Nb, Ta, Sb) O_3 [8–11]. Therefore, the structure and phase transition modulating is a key issue to design ceramic with high piezoelectric performance and stable temperature dependence [12–19].

In recent years, a remarkable discovery in phase transition adjusting of KNN-based ceramics has attracted the attraction of researches [20]. The sharp and narrow phase transition results in strong temperature dependence, whereas the broad and gentle phase transition causes to weak temperature dependence. And the “relaxor”-type transition contributes to the temperature stability improvement [20–22]. The relaxor behavior in KNN-based ceramics appears when at least two or three cations occupy the same

* Corresponding author.

E-mail address: yangwenlong1983@163.com (W. Yang).

crystallographic site A or B, which could be achieved by both A-site and B-site substitutions [22]. A-site cation disorder was always regarded as the main derivation of relaxor behavior [15,22]. The valence of A-site K, Na, and Li atoms in the perovskite structure are all +1, the ionic radius are 1.33 Å, 1.02 Å, and 0.68 Å, respectively. Ta and Nb are both located in the B-site with the +5 valence, the ionic radius are 0.64 Å and 0.65 Å, respectively. The La_2O_3 is used in this experiment as a doping addition, whose ionic form is La^{3+} and ionic radius is 1.06 Å. According to crystal chemistry and Goldschmidt Tolerance factor [20–22], La^{3+} might likely occupy in the A position. Therefore, the chemical formula of $(\text{Na}_{0.52}\text{K}_{0.44}\text{Li}_{0.04})_{1-3x}\text{La}_x\text{Ta}_{0.2}\text{Nb}_{0.8}\text{O}_3$ (KLNLTN-Lax) ceramics were determined in this paper based on chemical equilibrium. The diffuse phase transition was expected to appear in La-doped KLNLTN perovskite system, which aiming to improve the temperature stability of KNLTN ceramic by virtue of the diffuse phase transition behavior. Then, the crystalline phase and microtopography of KNLTN ceramics with different La doping concentrations were examined. The piezoelectric properties and temperature stability of KNLTN-Lax ceramic were investigated and the physical mechanisms that effects of La-doped on piezoelectric property and temperature stability of KNLTN ceramic in the view of valence compensation and polymorphic phase transition (PPT) were analyzed.

2. Experimental

$(\text{Na}_{0.52}\text{K}_{0.44}\text{Li}_{0.04})_{1-3x}\text{La}_x\text{Ta}_{0.2}\text{Nb}_{0.8}\text{O}_3$ (KLNLTN-Lax) ceramics ($x=0, 0.25, 0.5, 0.75, 1.00, 1.25$ mol%) were prepared by conventional ceramics sintering techniques, high-purity (99.99%) K_2CO_3 , Na_2CO_3 , Li_2CO_3 , Nb_2O_5 , Ta_2O_5 and La_2O_3 were used as raw materials. They were milled in polyethylene with agate balls for 24 h using ethanol as a medium. After calcination at 880 °C for 4 h, the calcined powders were ball milled again for 24 h, dried and then polyvinyl alcohol (PVA) was added as a binder. The sintering temperature was chosen from 1180 to 1100 °C for 4 h.

The phase structures of KNLTN ceramic with different La-doped concentrations were detected by X-ray powder diffraction (XRD-6000, Shimadzu, Japan). The surface and fracture topographies were observed with scanning electron microscope (SEM, S-4700, Hitachi, Japan). In order to study the piezoelectric property of material, the ceramic samples were polished, painted with silver, and polarized successively, and the polarization electric field was set as 4 kV/mm, the samples under the polarization electric field were heated to 120 °C in velocity of about 4 °C/min, then cooling down to room temperature after keeping 30 minutes, at last, the electric field was removed. The piezoelectric property of polarized samples was tested with quasi static piezoelectric tester (ZJ-3A) after aging 24 h.

3. Results and discussion

3.1. Structure of La-doped KNLTN ceramic

In order to confirm the lattice structure of La-doped KNLTN ceramic, the XRD spectra of different concentrations (0–1.25 mol%) La-doped KNLTN ceramics were measured, which were shown in Fig. 1. Pure perovskite structure without a second phase was presented when La doping concentrations were ranged from 0.00 mol% to 1.25 mol%, which indicate that the lattice tolerability was not obviously exceed. Different phase structures were obtained as La doped into KNLTN ceramics with different concentrations, KNLTN presented mixed tetragonal-orthogonal phase when La-doped concentrations are below 1.00 mol%. The variation

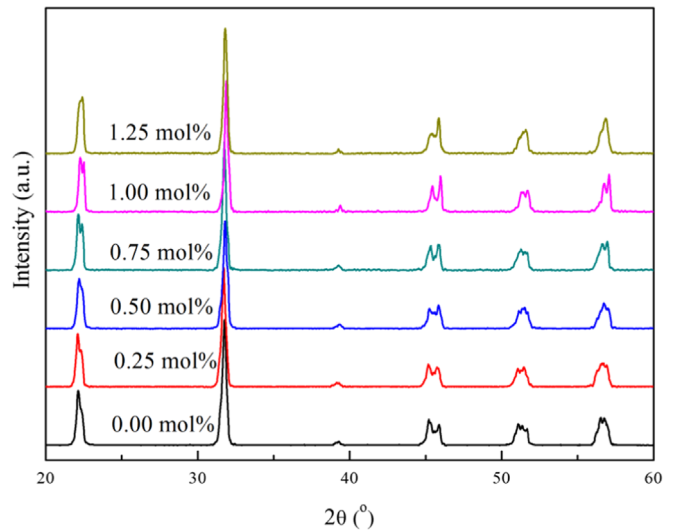


Fig. 1. XRD patterns of KNLTN ceramics with different content of La.

of the peak around 45° illustrates transition from mixed tetragonal-orthogonal phase to single tetragonal phase with the increasing La^{3+} doping concentration as shown in Fig. 1. Strictly speaking, pure tetragonal phase with P4mm space group were obtained when doping concentration is more than 1.00 mol%.

The surface and fracture microtopographies of La-doped KNLTN ceramics with 0.00 mol%, 0.75 mol% and 1.25 mol% La-doped concentrations were presented in Fig. 2. The surface microtopographies of KNLTN ceramics were shown in Fig. 2(a), (c) and (e), while the fracture microtopographies were shown in Fig. 2(b), (d) and (e), respectively. Grains shape and boundaries could be observed in the surface microtopographies of La-doped KNLTN ceramics obviously. The homogeneous grains size of pure KNLTN ceramics is about 6 μm. The grains size is about 2–4 μm when La doping concentration is 0.75 mol%, while the uniformity of grains degenerates due to the local 1–2 μm grains. Small crystalline grains about 1–2 μm have occupied more proportion when La doping concentration is 1.25 mol%. It can be seen obviously that the La element has significant effect on the grain size of KNLTN ceramics, and the grain growth could be restrained by La doping. The fracture micrographs of KNLTN ceramics show that all the ceramic samples present transgranular fracture, indicating that the binding force among crystalline grains is strong. Some micropores exist among the crystalline grains in the local part of ceramic samples. However, few micropores can be observed in the ceramics with 0.75 mol% La-doped KNLTN. And the grains boundaries of the grains are indistinctive compared with pure KNLTN ceramics, which indicate that binding force among the crystalline grains become strong as well. Many micropores could be found in the 1.25 mol% La-doped ceramics, which results in density degeneration and undesirable piezoelectric property. The increasing defects induced by chemical valence compensation in the KNLTN ceramic with excess La^{3+} were regarded as the main reason [20,23].

Therefore, the La doping has significant influences on microtopography structure and sintering property. It not only can reduce the sintering temperature of ceramic as sintering addition, but also restrain the growth of crystalline grains. Appropriate concentration La-doping will contribute to decreasing the numbers of micropores and excellent density can be achieved. In this paper, high density of 4.72 g/cm³ and relative density of 94.4% of pure KNLTN ceramic was obtained. The densities increase with increasing La-doping, and the values will achieve to maximum 4.77 g/cm³ and 95.4% in 0.75 mol% La-doped ceramics as shown in Fig. 3. It is not

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