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ScienceDirect
Ceramics International 40 (2014) 4341–4344
CERAMICS INTERNATIONAL
www.elsevier.com/locate/ceramint

Enhanced piezoelectric properties of $(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$ lead-free ceramics by optimizing the sintering temperature

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Received 19 August 2013; received in revised form 23 August 2013; accepted 23 August 2013
Available online 30 August 2013

Abstract

Effects of sintering temperature on the microstructure and electrical properties of $(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$ (KNLNS) lead-free ceramics are investigated. The grain size gradually becomes larger with increasing sintering temperature from 1055 °C to 1105 °C, and the piezoelectric property could be enhanced by optimizing their sintering temperature. The ceramic sintered at 1075 °C has optimum electrical properties, i.e., $d_{33}\sim 272$ pC/N, $k_p\sim 43.5\%$, $\varepsilon_r\sim 1152$, tan $\delta\sim 0.026$, and $T_C\sim 346$ °C. These results show that the sintering temperature can optimize electrical properties of KNLNS ceramics.

Keywords: C. Electrical properties; C. Piezoelectric properties; $(KNa)NbO_3$; Lead-free ceramics; Sintering temperature

1. Introduction

Lead-based piezoceramics have been widely used for piezoelectric actuators, sensors, and transducers because of their excellent electrical properties [1]. However, the lead of lead-based ceramics has resulted in the environmental pollution during their preparation and process [1]. Therefore, lead-free piezoceramics have been extensively investigated for protecting the living environment, the material system including barium titanate, bismuth titanate, and alkali niobate [2–23]. Among those, considerable attention has been given to (K,Na)NbO_3 (KNN) lead-free piezoceramics because of their good piezoelectric properties and high Curie temperature [6–23].

Some efforts have been made to improve the d_{33} of KNN-based ceramics [6–23], such as ion substitution, sintering aids, and different preparation methods. It is well known that the ion substitution can improve the piezoelectric properties of KNN [6–18]. Enhanced piezoelectric properties of 212–280 pC/N have been observed in Li^+ and Sb^{5+}-modified KNN ceramics due to the involvement of the orthorhombic–tetragonal phase boundary [9–18]. For example, Wu et al. reported an enhanced d_{33} of 262–280 pC/N in Li^+ and Sb^{5+}-doped KNN ceramics [9,12,13]. Palei et al. reported that $LiSbO_3$-modified KNN ceramics have a d_{33} of $\sim$215 pC/N [14]. These results shows that piezoelectric properties of Li^+ and Sb^{5+}-doped KNN ceramics are sensitive to the preparation condition [9–18]. In addition, the sintering temperature is a key factor to determine the electrical properties of KNN-based ceramics [24,25]. Zhao et al. reported that the piezoelectric coefficient of $\sim$268 pC/N can be achieved by optimizing the sintering temperature of $LiTaO_3$-doped KNN ceramics [25]. However, there are few reports on the effect of the sintering temperature on the microstructure and piezoelectric properties of $(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$ lead-free ceramics.

In this work, we studied the effect of the sintering temperature on the microstructure and electrical properties of $(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$ (KNLNS) lead-free ceramics prepared by the conventional solid-state method. Enhanced

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http://dx.doi.org/10.1016/j.ceramint.2013.08.102

piezoelectric properties of $d_{33}\sim 272$ pC/N and $k_p\sim 43.5\%$ are achieved by optimizing the sintering temperature. Underlying physical mechanisms for enhanced piezoelectric properties were also discussed.

2. Experimental procedure

$(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$ lead-free piezoceramics were prepared by the conventional solid-state method. Raw materials of Li_2CO_3 (99%), Nb_2O_5 (99.5%), Sb_2O_3 (98%), K_2CO_3 (99%), and Na_2CO_3 (99%) were weighed according to the stoichiometric formula of $(K_{0.40}Na_{0.60})_{0.94}Li_{0.06}Nb_{0.94}SbO_3$, and were then mixed for 24 h by the ZrO_2 balls using the ethanol as the medium. These powders were dried and calcined at 800 °C for 4 h, and then these sintered powders were pressed into disks of 1.0 cm in diameter and 1.0–1.2 mm in thickness under 15 MPa. These green pellets were sintered at the temperature range of 1055–1105 °C for 3 h. These sintered pellets were polished and then coated with the silver paste on both surfaces as the electrode for the electrical measurement, and were polarized in the silicone oil of 50 °C under an electric field of 4.0 kV/mm for 20 min.

The crystal structure of these sintered ceramics was measured using the X-ray diffraction (XRD) with Cu-K_a radiation, and their surface morphologies were detected using the scanning electron microscope. The density of the sintered samples was measured by the Archimedes method. Their dielectric behavior as a function of the measurement temperature was obtained using an *LCR* meter (HP 4980, Agilent, USA). Their piezoelectric constant (d_{33}) was characterized using the quasi-static piezoelectric d_{33} meter (ZJ-3A, China), and the planar coupling factor (k_p) was calculated using the resonance and anti-resonance method.

3. Results and discussion

Fig. 1 shows the XRD patterns of KNLNS ceramics sintered at different temperatures of 1055–1105 °C, and all the data was collected at room temperature. All ceramics have a pure perovskite structure, and secondary phases cannot be detected in the measurement range of the XRD machine. As a result, the sintering temperature of 1055–1105 °C does not change the phase structure of KNLNS ceramics.

Fig. 2(a–c) shows the SEM patterns of KNLNS ceramics sintered at different temperatures. The grain size becomes larger as the sintering temperature increases. It was found that some large grains with > 10 μm exist in the ceramics sintered at a higher temperature of ≥ 1075 °C, as shown in Fig. 2(b) and (c). In addition, some small grains locate at the gaps among the larger ones, giving rise to a dense microstructure [Fig. 2(b)]. In order to confirm the effect of the sintering temperature on their density, the relative density is given in Fig. 3. It was observed that the optimal sintering temperature results in the denser microstructure, that is, the ceramics sintered at 1065–1085 °C have a higher density. It was reported that the low sintering temperature results in the formation of some pores, thus a lower density is formed in the KNN-based ceramics [24]. However, a high processing temperature easily causes the loss of Na and K [25,26], showing a poor density. As a result, the optimal sintering temperature causes the denser microstructure of KNLNS ceramics [24–26].

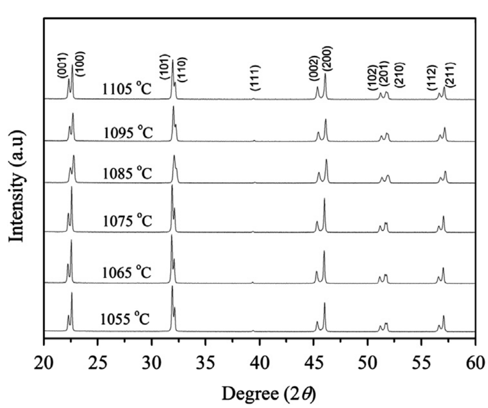

Fig. 1. XRD patterns of KNLNS ceramics sintered at different temperatures.

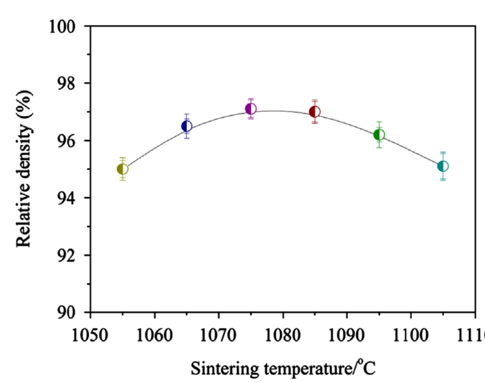

Fig. 3. Relative density of KNLNS ceramics as a function of the sintering temperature.

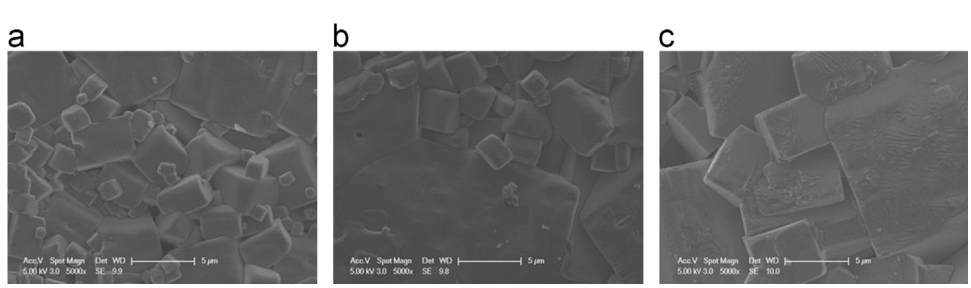

Fig. 2. Surface morphologies of KNLNS ceramics sintered at (a) 1055 °C, (b) 1075 °C, and (c) 1105 °C.