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Short communication

Effect of sintering temperature on ferroelectric properties of $0.94(K_{0.5}Na_{0.5})NbO_3$–$0.06LNbO_3$ system

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Abstract

Lead free $0.94(K_{0.5}Na_{0.5}NbO_3)$–$0.06(LiNbO_3)$ (KNN–LN) system was synthesized by conventional solid state reaction route (CSSRR). The KNN–LN system was calcined at 850 °C for 6 h for the formation of single perovskite phase whereas the sintering was done at 1050 °C, 1080 °C and 1100 °C for 4 h, respectively. The KNN–LN samples sintered at 1080 °C show better properties: room temperature (RT) dielectric constant (ε_r) ~936, dielectric loss (tan δ) ~0.016 at 1 MHz, a relatively high bulk density (ρ) ~4.385 g/cm^3, which is 97.5% of the theoretical density (TD ~ 4.51), remnant polarization (P_r) ~6.4 μC/cm^2 and coercive field (E_c) ~9.6 kV/cm have been observed.

Keywords: A. Powders: solid state reaction; A. Sintering; C. Dielectric properties; C. Ferroelectric properties

1. Introduction

Ferroelectric ceramics are the heart and soul of several multibillion dollar industries, ranging from high-dielectric-constant capacitors to later developments in piezoelectric transducers, positive temperature coefficient devices and electro-optic light valves [1]. Among lead oxide-based ferroelectrics, lead zirconate titanate (PZT) system was intensively studied for many years [2]. A noticeable feature of PZT material is the occurrence of a morphotropic phase boundary (MPB) [2–5]. The compositions close to the MPB show excellent piezoelectric and ferroelectric properties and hence, they have been widely used for piezoelectric actuators, sensors and transducers applications [6]. However, the high toxicity of lead oxide has caused serious environmental problems. Therefore, investigations have been extensively carried out to develop lead free systems with potentially good piezoelectric and ferroelectric properties [7].

Lead free ferroelectric materials with perovskite structure have attracted much attention since they are easy to prepare and the structure is simple compared to other structures [1]. Many lead free ferroelectric materials such as barium titanate (BT), bismuth sodium titanate (BNT), potassium niobate (KN), potassium sodium niobate (KNN) and potassium tantalate niobate (KTN) have a perovskite structure [8,9]. Among all the lead free ferroelectric materials $(K_{0.5}Na_{0.5})NbO_3$ (KNN) based systems have attracted much attention because of having high piezoelectric properties, high Curie temperature (T_c) and environmental friendly nature [10–19]. A major problem concerning KNN system is difficulty in obtaining high density by conventional solid state reaction route (CSSRR) for several reasons [20,21]. One is that for KNN phase stability is limited to 1140 °C [22]. In addition, a deviation from stoichiometry can result in the formation of extra phases [23]. Therefore, in order to solve the above problems, various synthesis techniques have been utilized, such as hot pressing and spark plasma sintering process [24,25]. Although these methods can yield high densities and better properties compared to KNN synthesized by CSSRR, but still careful investigation and optimization of sintering parameters are needed to obtain reproducible and high quality materials. Additionally, since they are more expensive routes compared to the CSSRR, efforts have been made to find new systems based on KNN that can be synthesized by CSSRR with improved electrical properties.

In this work, the lead free KNN–LN system has been synthesized by conventional solid state reaction route. The

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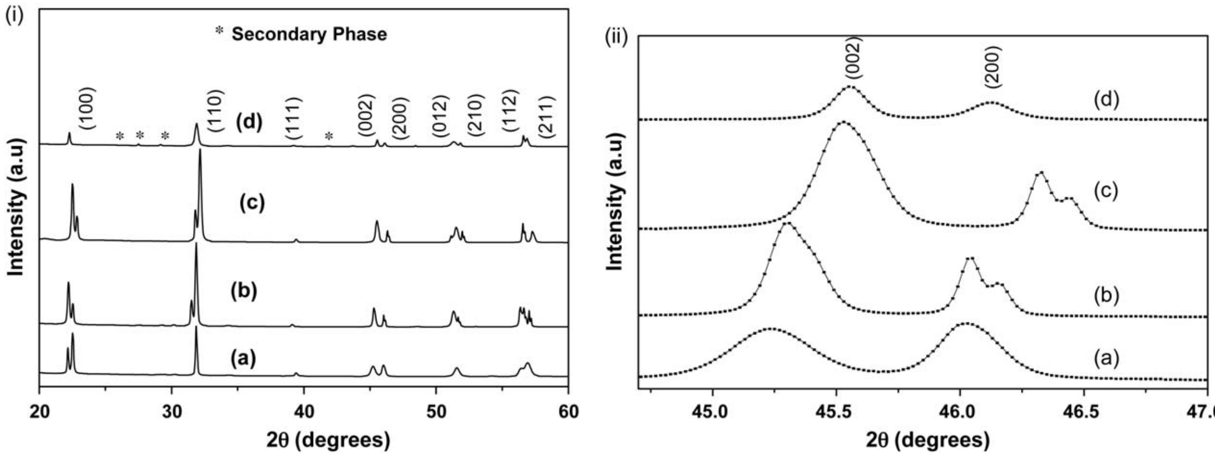

Fig. 1. (i) XRD patterns of KNN–LN system calcined at (a) 850 °C, sintered at (b) 1050 °C. (c) 1080 °C and (d) 1100 °C. (ii) Enlarged XRD pattern.

effects of sintering temperature on density, remnant polarization and dielectric properties have been studied.

2. Experimental procedure

Lead free KNN–LN ceramics were prepared by a conventional solid state reaction route. Sodium carbonate (Na_2CO_3, 99% purity), potassium carbonate (K_2CO_3, 99% purity), lithium carbonate (Li_2CO_3, 99% purity), and niobium pentoxide (Nb_2O_5, 99% purity) were used as starting materials. Stoichiometric weights of all the powders were mixed and ball milled with acetone for 8 h, using zirconia balls as the grinding media. After drying the slurry in oven, the calcination was carried out at 850 °C for 6 h and single phase formation was confirmed by the X-ray diffraction (XRD) technique. The calcined mixture was mixed thoroughly with 2 wt% polyvinyl alcohol (PVA) binder solution and then pressed into disks of diameter of 10 mm and a thickness of 1.5 mm under ~60 MPa pressure. The sintering of the samples was carried out at 1050 °C, 1080 °C and 1100 °C for 4 h, respectively with a

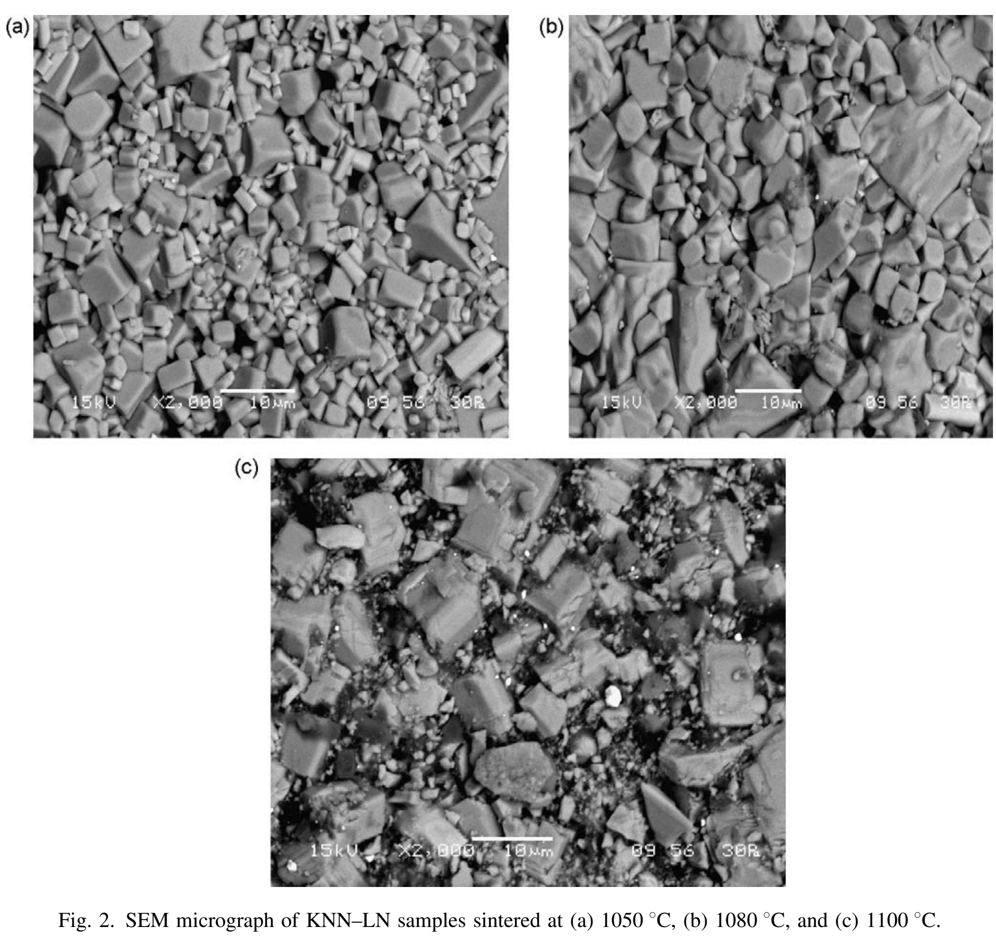

Fig. 2. SEM micrograph of KNN–LN samples sintered at (a) 1050 °C, (b) 1080 °C, and (c) 1100 °C.