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Preparation, microstructure and relaxor ferroelectric characteristics of BLNT–BCT lead-free piezoceramics

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ABSTRACT

In the present study, lead-free piezoelectric (1-x)[$(Bi_{0.96}La_{0.04})_{0.5}Na_{0.5}TiO_3$]-(x)[$Ba_{0.90}Ca_{0.10}TiO_3$] ceramics with $x \leq 0.20$ have been prepared at optimum temperature (950°C–1050 °C) by employing semi-wet technique. The effect of $Ba_{0.90}Ca_{0.10}TiO_3$ (BCT) content on the phase formation, surface morphology, dielectric and piezoelectric properties is systematically investigated and characterized by X-ray diffraction (XRD), Raman Spectroscopy, Field emission Scanning electron micrographs (FE-SEM), Impedance analyzer. XRD analysis shows phase transformation from rhombhohedral to tetragonal with the addition of BCT and the average grain size decreases with BCT content, x. Temperature dependent dielectric behavior exhibits two phase transitions, i.e. depolarization temperature; T_d and phase transition temperature; T_m. The phase transitions; T_m tends to diffuse with increasing x and exhibited a broad frequency dependent maximum upto $x \leq 12$. Optimum piezoelectric and dielectric properties (i.e., $d_{33} = 165$ pC/N, $k_p = 39\%$, $k_t = 47\%$ and $k_{33} = 57\%$, $\varepsilon_r = 1585$, tan $\delta = 0.07$) in the studied system have been obtained.

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1. Introduction

Lead-free piezoelectric ceramics have received global interest as an alternative of PZT based ceramics. As, PZT based piezoelectric ceramics are widely used in high performance sensors and hysteresis-free actuators due to their outstanding dielectric and piezoelectric properties [1–4]. These materials contain large amount of lead (≥60%), which is toxic in nature and it can cause damage to the brain, kidney and nervous system, specially the intelligence of the children [5,6]. So, there is a great demand to extend lead free piezoelectric materials whose properties are comparable to lead based systems. In the family of lead free material systems, there are a few bismuth based materials like BNT and BKT apart from non-bismuth based materials like KNN, BT, BZT etc. [7,8] which have a potential to replace lead-based materials from the electronic industry.

BNT was first discovered by Smolenskii et al. [9] and well-known to be a promising material for lead free piezoelectric applications owing to its high Curie temperature (320 °C). Pure BNT ceramic exhibits ferroelectric properties because of the presence of stereochemically active lone-pair electrons of Bi [10]. Although, the inadequacy of this material is its high conductivity and high coercive field ($E_c = 7.3$ kV/mm) [11]. As for as piezoelectric properties are concerned, it is much inferior to that of $Pb(Zr_{0.52}Ti_{0.48})O_3$ ceramics [12]. To solve these problems along with improvement in the electrical properties, varieties of compositional modifications have been made into the BNT system to form solid solutions. In recent years, BNT based solid solutions have received great attention, mainly because of its fascinating electromechanical properties at the structural phase boundary between the two/three end members [11,13–18]. In our previous work, rare earth modified BNT has shown very interesting dielectric and piezoelectric properties [19–21]. Moreover, Pengpat et al. (2006) found that La_2O_3 addition on BNT-BT system show remarkable increment in piezoelectric coefficient (d_{33}) [22] On the other hand, Barium calcium titanate (Ba1-xCaxTiO3) solid solution has attracted great interest and being used in optical storage devices, advanced laser systems, and capacitor materials including multilayer ceramic capacitor, dielectric filter, dielectric antenna, dielectric resonators and piezoelectric

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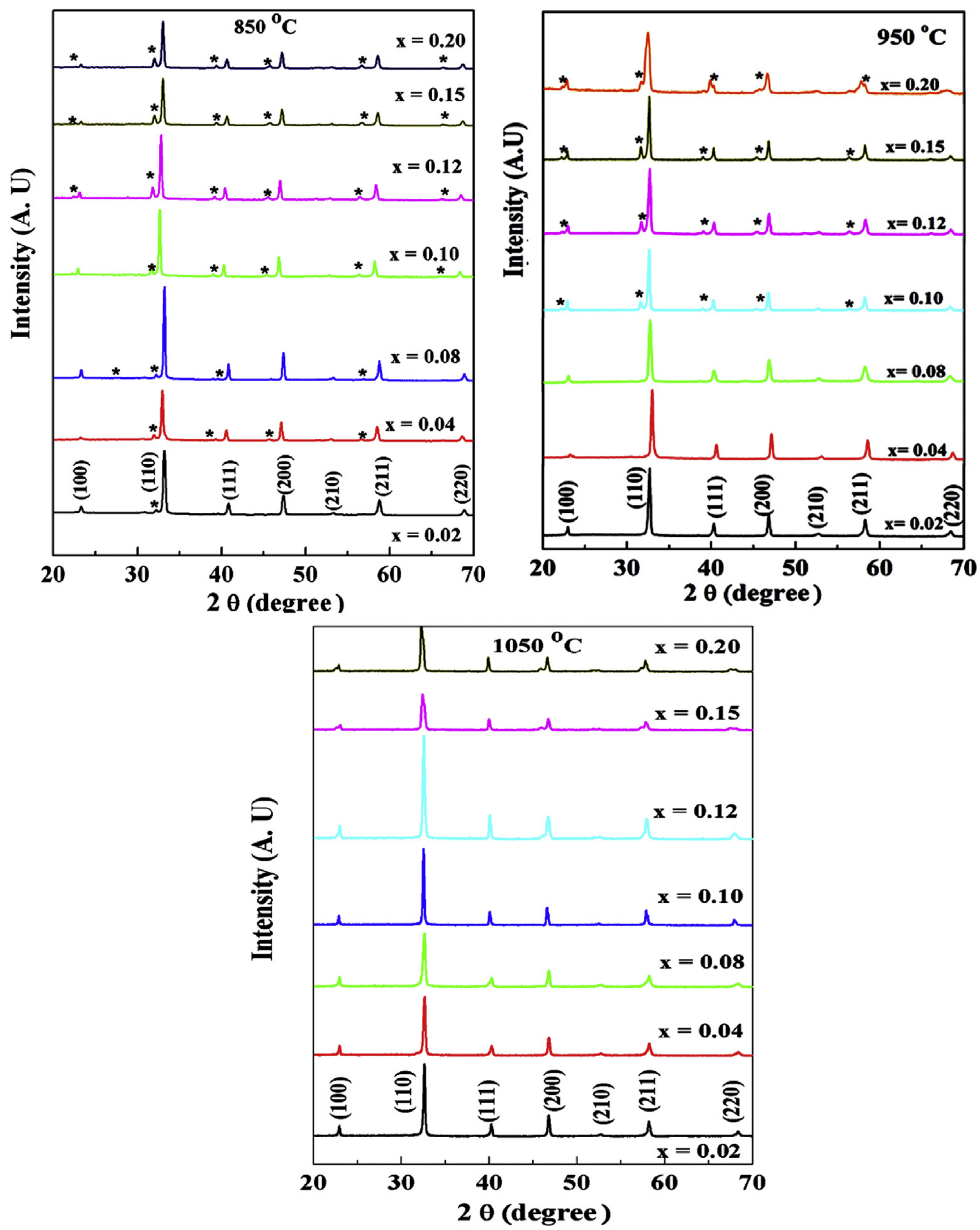

Fig. 1. XRD patterns of 1−xBLNT-xBCT ceramics at different temperature (a) 850 °C, (b) 950 °C and (c) 1050 °C.

actuators [23,24].

Based on the unique characteristics of BNT and BCT system, we have chosen to study the solid solution formation of binary (1-x) BLNT-(x)BCT ceramics by semi wet technique and to investigate their phase formation, dielectric, piezoelectric and electromechanical behavior in details. To the best of our knowledge, there is no report available on dielectric, piezoelectric and eletromechanical properties of (1-x)BLNT-(x)BCT system for x upto 0.20.

2. Experimental details

The compositions of the (1-x)BLNT-(x)BCT ceramic system with $x \leq 0.20$ were synthesized by semi wet technique. High purity AR-grade metal oxides or nitrate powders (Sigma Aldrich) were used as raw materials which are Bi_2O_3 (99%), $NaNO_3$ (99%), La_2O_3 (99.9%), $Ba(NO_3)_2$ (99.9%), $Ca(NO_3)_2 \cdot 4H_2O \cdot$ (99.9%), TiO_2 (99.9%), and ethylene glycol. In order to substitute La^{3+} on A-sites rather than B-sites in BLNT ceramics and simultaneously Ca^{2+} on A-sites rather than B-sites of BCT ceramics, BLNT and BCT ceramics were separately synthesized by semi wet technique. The detail of synthesis process was described elsewhere [21]. The BLNT and BCT ceramics were individually calcined at 850 °C (2 h) and 1250 °C (10 h) respectively. For single phase formation, the mixed powders of all (1-x)BLNT-xBCT ceramic compositions were calcined at three different temperatures 850 °C, 950 °C and 1050 °C for 4 h. After calcination, powders were reground and mixed with organic binder (PVA) followed by pelletization in the form of cylindrical disc of dia. 10 mm using uni-axial hydraulic press. These pellets were kept at 500 °C for 4 h to burn off the organic binder and were sintered at three different temperatures (1100 °C, 1150 °C and 1200 °C for 2 h) in air atmosphere. Thereafter, two pellets of each composition of higher density were coated with silver paste on both sides and cured at 400 °C for 20 min for the subsequent electrical measurements.

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