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Research Article

Dielectric and Ferroelectric Properties of Lead-Free $[1-z\{(Bi_{1-x}La_x)_{0.5}(Na_{1-y}Li_y)_{0.5}TiO_3\}\text{-}zBaTiO_3]$ Ceramic System

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Lead-free specimens of $[1-z\{(Bi_{1-x}La_x)_{0.5}(Na_{1-y}Li_y)_{0.5}TiO_3\}\text{-}zBaTiO_3]$ (BLNLT-BT, where $x = 0.04$, $y = 0.025$, and $z = 0$, 0.02, 0.04, and 0.06) ceramic system were synthesized by a semiwet technique. In the present study, we have investigated the effect of Ba on the structure, morphology, and dielectric, ferroelectric, and piezoelectric properties of BLNLT system. The XRD patterns of all the specimens showed rhombohedral structure at room temperature. Field emission scanning electron microscopy images (FE-SEM) showed grain growth inhibition with Ba content in BLNLT system. The temperature dependence of dielectric constant revealed that the temperature of maximum dielectric constant "T_m" and depolarization temperature "T_d" decreased for the sample $z = 0.06$ and showed high value of dielectric constant with low dielectric loss. These samples exhibited nonlinear behaviour of hysteresis loop. The values of dielectric constant (ε_r), dissipation factor ($\tan\delta$), piezo charge coefficient (d_{33}), remnant polarization (P_r), and coercive field (E_c) for the composition $z = 0.06$ were 1495, 0.06, 20 pC/N, 5 μC/cm^2, and 10.5 kV/cm, respectively.

1. Introduction

Lead-free piezoelectric ceramics were studied extensively over the last few decades due to environmental concern as well as government regulations against the use of hazardous substances [1]. Bismuth sodium titanate, $Bi_{0.5}Na_{0.5}TiO_3$ (BNT), was considered to be a promising lead-free material, and discovered by Smolenskii et al. in 1961 [2]. BNT is a ferroelectric material having Bi^{3+} and Na^+ ions on the A-sites of ABO_3 type perovskite structure with a rhombohedral symmetry. It has high Curie temperature (T_c ~ 320°C) and strong ferroelectric nature at room temperature [3]. However, the shortcoming of this material is its high conductivity and high coercive field (E_c = 7.3 kV/mm) [4, 5]. Therefore, pure BNT ceramic usually exhibits weak piezoelectric properties. Among the studied materials, solid solutions between a rhombohedral $(Bi_{1/2}Na_{1/2})TiO_3$ (BNT) and a tetragonal $BaTiO_3$ (BT) have been of particular interest largely because of morphotropic phase boundary (MPB). A morphotropic phase boundary (MPB) exists at 6-7 mol% BT. The BNT-BT system was firstly reported by Takenaka et al. [6]. To improve the electrical properties of material, a number of BNT-based ceramics have been usually produced by the convectional solid state method and various chemical methods, such as hydrothermal process [7], citrate method [8], sol-gel, and autocombustion [9]. Recently, a lot of efforts have been made to improve their densification behaviour and electrical properties through doping elements [10, 11] and few studies revealed that there were many possibilities to further enhance the piezoelectric properties of KNN-based and Bi-based ceramics even through simple Li doping [12–14]. Due to volatile nature of Bi_2O_3 at high sintering temperature (>1200), Li was often used in BLNT compositions to achieve enhanced dielectric, ferroelectric, and piezoelectric properties [15]. In order to get highly dense ceramic, this sample was sintered at 1200°C for 2 hours. To avoid the volatility of Bi_2O_3 and getting high density at low temperature, we have substituted La/Li (x/y) on A-site in BNT system. As we know that La acts as modifier and enhances the electrical properties [16] and Li improves the densification with reduced sintering temperature [15]. In the present work, we demonstrate the influence on crystal structure, dielectric, ferroelectric, and piezoelectric properties of Ba substitution in $1-z$(BLNLT)-z(BT) system where $z = 0$, 0.02, 0.04, and 0.06. To the best of our knowledge, the specimens of [$1-z$(BLNLT)-z(BT) system, $x/y/z = 0.04/0.025/0$–0.06] were synthesized and studied in detail for the first time using semiwet technique. The potential advantages of semiwet technique over sol-gel and solid state reaction technique are controlled size and shape, atomic level homogeneity of doping elements at A-site and B-site in ABO_3 perovskite structure and it is a promising technique to produce fine multicomponent oxide ceramic powders in a very simple and economic way. This technique was found to be very useful in improving the properties [17].

2. Experimental Procedure

Lead-free specimens of $1-z$(BLNLT)-z(BT), $x/y/z$ = 0.04/0.025/0–0.06 were prepared by semi-wet technique using AR-grade metal oxides or nitrate powders (Sigma Aldrich) as raw materials like Bi_2O_3 (99%), $NaNO_3$ (99%), La_2O_3 (99.9%), $LiNO_3$ (99.9%), TiO_2 (99.9%), and ethylene glycol. All the raw materials were weighed in appropriate amount according to their chemical compositions. The BLNLT and BT compositions were synthesized separately. The A-site of BLNLT system was prepared by using ethylene glycol precursor solution, in which ethylene glycol was expected to distribute the cations in atomic level forming a polymeric complex which was combusted at appropriate temperature ($T \sim 150$–200°C) in the form of ash powders. The ash, highly fine, homogeneous, and highly reactive, was mixed thoroughly with appropriate amount of TiO_2 powder in ethanol media for 2 hrs. The details of synthesis process were described elsewhere [18]. Dried powders for these compositions were calcined at 750°C for 2 hours. In order to get highly dense ceramic, sintering was done at 1100°C for 2 hours. To avoid the volatility of Bi_2O_3 and getting high density at low sintering temperature, we had substituted La/Li (x/y) on A-site in BNT system separately. Thereafter, the specimens of [$1-z$(BLNLT)-z(BT) complex system with compositions 0.04/0.025/0–0.06] were synthesized by semiwet method and calcined at temperature that is, 800–900°C for 2 hours. After calcination, powder were reground with binder (polyvinyl alcohol) and pressed into pellets of diameter 10 mm and thickness ≤1.5 mm using hydraulic press. These pellets were kept at 400°C for 4 hours to burn off the binder, and temperature was raised to 1100–1150°C and kept at this temperature for 2 hours for sintering.

The crystalline structure of the sintered samples was examined using X-ray diffraction analysis with Cu-Kα radiation (XRD-6000, Shimadzu, Japan). The surface morphology of sintered samples was studies by field emission scanning electron microscopy (FE-SEM, Quanta 200 FEG, FEI, Netherlands). Bulk density of the sintered sample was measured by the Archimedes method. The relative density was determined using bulk density (calculated by taking mass and dimensions of the pellet) and theoretical density (determined using XRD data). Silver paste was coated on both sides of these sintered and polished samples as electrodes for the purpose of electrical measurements, and cured at 500°C for

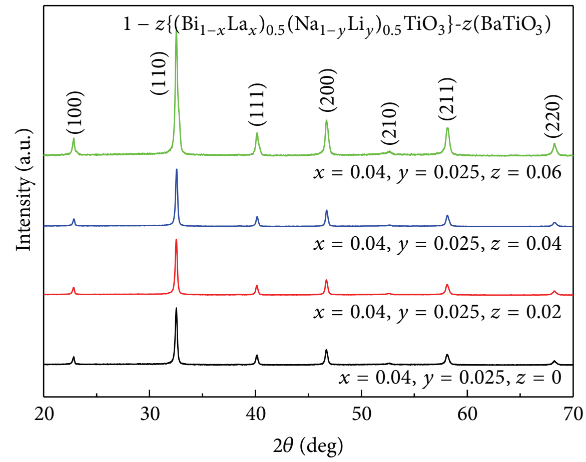
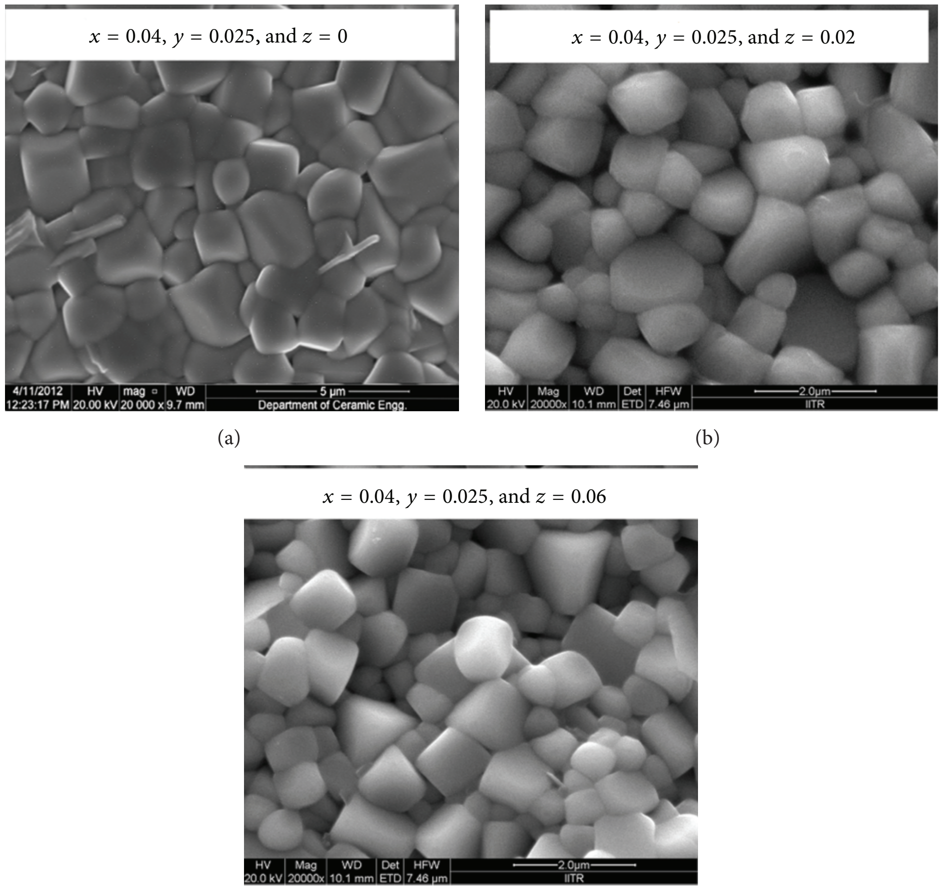


Figure 1: XRD patterns of [$1-z$(BLNLT)-z(BT), $x/y/z$ = 0.04/0.025/0, 0.02, 0.04, 0.06] system.

20 min. The dielectric constant (ε_r) and dielectric loss ($\tan\delta$) of these samples at 1 kHz, 10 kHz, and 100 kHz were measured as a function of temperature over the temperature range from room temperature to 500°C using LCR meter (Hioki 3522); P-E loop tracer (Marine India), based on modified Sawyer-Tower circuit, was used to trace polarization versus electric field loops at 50 Hz. Piezoelectric charge constant (d_{33}) was measured after poling using d_{33} Piezometer (Take Control, PM 35) for all the specimens. Poling was done under 3 kV/mm DC electric field at 50°C for 1 hour duration.

3. Results and Discussion

Figure 1 shows the X-ray diffraction pattern of lead free [$1-z$(BLNLT)-z(BT), $x/y/z = 0.04/0.025/0$–0.06] system. It was observed from the diffraction patterns that all the specimens exhibited single-phase formation with rhombohedral structure and no secondary phase(s) were observed within detection limit of XRD. It implied that La^{3+}, Li^{+}, and Ba^{2+} have diffused into BNT lattice to form a solid solution. Even though many investigations have been carried out on the symmetry of BNT-based ceramics; yet, there is still controversy in crystallographic symmetry. A large number of researcher reported that BNT has rhombohedral structure [19–21]. However, few reports suggested that the BNT has a lower symmetry with monoclinic phase [22, 23]. In the present system, we found that the symmetry of the solid solution became more complex for BLNLT-BT solid solution. In order to confirm the structure and symmetry, further study had been carried out through some structural software, which will be presented later. The bulk density of the sintered specimens was measured by the Archimedes method. The relative density was determined using bulk density (calculated by taking mass and dimensions of the pellet) and theoretical density (determined using XRD data). The bulk density observed in [$1-z$(BLNLT)-z(BT), $x/y/z = 0.04/0.025/0$] system was 5.8 gm/cc which is 97% of theoretical density, and the bulk density observed in [$1-z$(BLNLT)-z(BT),



(a) (b) (c)

Figure 2: Microphotographs of [1 − z(BLNLT)-z(BT)] system for z = 0, 0.02, and 0.06.

Table 1: Electrical properties of [1 − z BLNLT-zBT] system.

Compositions $x/y/z$ x = 0.04, y = 0.025	At RT (100 kHz) ε_r	Tan δ	T_d (°C)	T_m (°C)	γ	D_{33} (pC/N)	D_{av} (μm)	P_r (μC/cm^2)	E_c (kV/cm)
z = 0	1365	0.08	170	355	1.69	70	2.67	29.26	28.46
z = 0.02	1386	0.27	130	305	1.58	65	2.34	10.29	15.35
z = 0.04	1423	0.08	120	290	1.66	47	—	5.96	15.16
z = 0.06	1494	0.06	80	225	1.79	20	1.78	4.87	10.59

$x/y/z$ = 0.04/0.025/0.06] system was 5.7 gm/cc which is 95% of theoretical density.

Figure 2 showed microstructure of all the sintered samples of [1 − z(BLNLT)-z(BT), $x/y/z$ = 0.04/0.025/0, 0.04/0.025/0.02 & 0.04/0.025/0.06] system taken for polished and etched surfaces. The microstructures of all the sintered samples were homogeneous, dense, and pore free. From the micrographs, it was evident that average grain size was found to decrease with Ba substitution in BLNLT system, which was consistent with the reported results [24]. In the present system, grain growth was slightly inhibited with the substitution of Ba^{2+} and small grains were formed, which may be due to the reduction in the mobility of the grain boundary. Thus, the mass transportation was weakend after the addition of Ba^{2+} [25]. The linear intercept method was used to determine the average grain size (D_{av}) of these samples (See Table 1).

Figure 3 illustrated the temperature dependence of dielectric constant and dielectric loss of BLNLT-BT ceramics at different frequencies (1, 10, and 100 kHz). The temperature dependence of ε_r showed that ε_r increased upto certain temperature and exhibited broad dielectric maxima around T_m, which decreased gradually with further increase in temperature above T_m. In general, two broad dielectric anomalies were obtained in BNT system, which is known to be T_d and T_m where "T_d" referred as depolarization temperature, corresponding the transition from ferroelectric to antiferroelectric transition, and it can also be derived from the peak

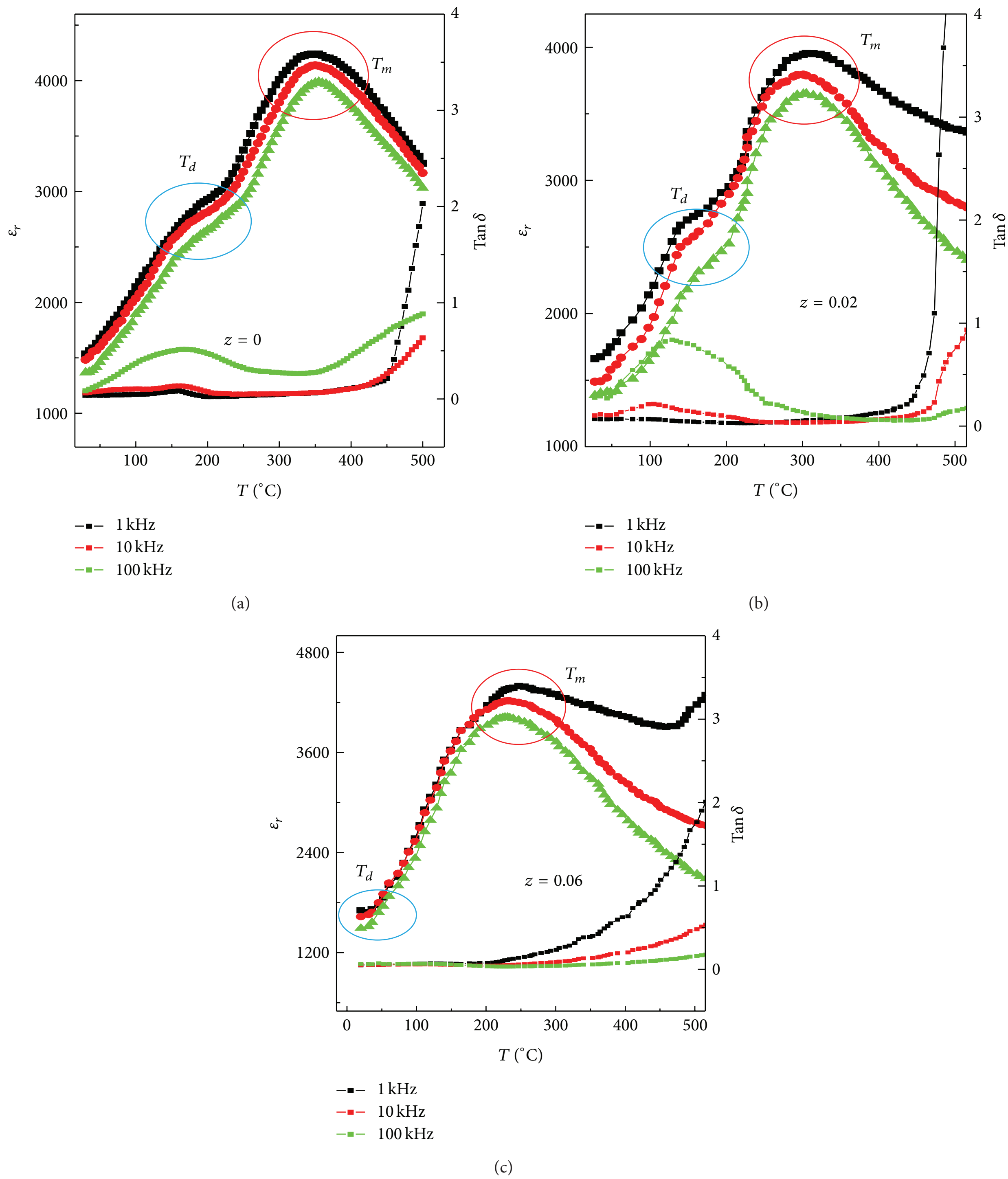


Figure 3: The temperature dependence of ε_r and tan δ of [1 − z (BLNLT)-z(BT)] system for samples z = 0, 0.02, and 0.06 at 1 kHz, 10 kHz, and 100 kHz.

in the temperature dependence plot of tan δ [26], and "T_m" referred as the temperature of maximum dielectric constant which corresponds to the transition from antiferroelectric to paraelectric phase transition. It was observed that dielectric plots of specimen $(x/y/z)$ = (0.04/0.025/0) exhibited slight frequency dependence above T_d (near T_m) and for specimens $(x/y/z)$ = (0.04/0.025/0.02–0.06), the dielectric constant was frequency dependent especially at low frequency and at higher temperature, and it seemed to be common feature in ferroelectric materials associated with ionic conductivity, and generally referred as low frequency dielectric dispersion [27]. The relative effect of ionic conductivity became small with increasing frequency. As a result, the frequency dependence of ε_r becomes weak. However, tan δ showed variation

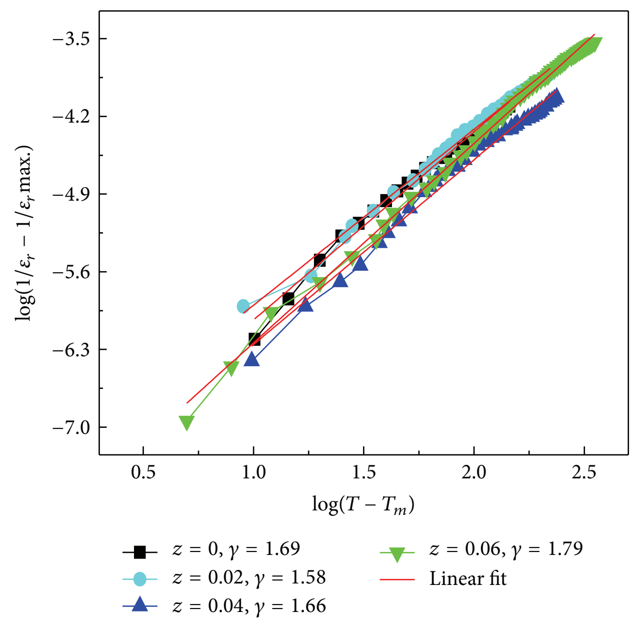


Figure 4: The $\text{Log}(1/\varepsilon_r - 1/\varepsilon_{r\,\max})$ versus $\log(T - T_m)$ of [1 − z(BLNLT)-z(BT), $x/y/z$ = 0.04/0.025/0–0.06] system at 100 kHz.

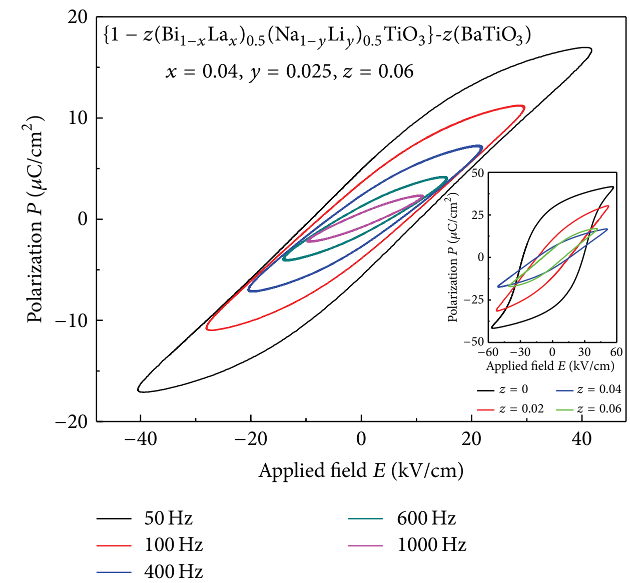


Figure 5: The P-E hysteresis loops of [1 − z(BLNLT)-z(BT), $x/y/z$ = 0.04/0.025/0.06] ceramics at RT.

with frequency in a different manner, $\tan\delta$ increased with increasing frequency and the increment was ascribed to the ionic conductivity. The retardation in polarization caused from ionic conductivity was enhanced with an increase in frequency that led to an increase of $\tan\delta$ [28, 29], Figures 3(b) and 3(c). The dielectric constant decreased whereas T_m slightly increased with increase in frequency [30, 31].

As evident from Figure 3, the values of dielectric constant and dielectric loss were decreased at higher frequencies, with the addition of Ba in BLNLT system. It was observed that the phase transition at T_m is a diffuse type phase transition as there was no dispersion between T_m versus frequency in the BLNLT-BT system. The phase transitions T_d (blue circle) and T_m (red circle) shifted towards low temperature with increasing Ba content in BLNLT system (see Figure 3), which was expected owing to low Curie temperature (T_c ~ 130°C as compared with that of BLNT ~ 355°C) of Barium titanate. The maximum dielectric constant and low value of dielectric loss were observed for the specimen $x/y/z$ = 0.04/0.025/0.06 at room temperature. For all the specimens, the values of dielectric constant and dielectric loss were listed in Table 1.

The diffuseness of BLNLT- BT ceramics can be expressed by equation, proposed by modified Curie-Weiss law [32],

$$\frac{1}{\varepsilon_r} - \frac{1}{\varepsilon_{r\,\max}} = C^{-\gamma}(T - T_m)^{\gamma}, \quad (1)$$

where, C is Curie coefficient, γ is degree of diffuseness, and $\varepsilon_{r\,\max}$ the maximum value of dielectric constant at T_m. The exponent "γ" can have a value ranging from 1 (for normal ferroelectric) to 2 (for an ideal relaxor ferroelectric). A linear relationship was observed in all the samples above T_m from $\ln(1/\varepsilon_r - 1/\varepsilon_{r\,\max})$ versus $\ln(T - T_m)$ plots, which is shown in Figure 4. For present system, the values of "γ" lie in the range of 1.58–1.79, which indicated that the material is highly disordered and diffuse phase transition was observed in BLNLT-BT system. The diffuseness was attributed mainly to the structural disordering and compositional fluctuations in the arrangement of cations in one or more crystallographic sites of the structure.

Figure 5 showed the room temperature hysteresis (P-E) loops for the sample $x/y/z$ = 0.04/0.025/0.06 at different frequencies. It was found that the sample with $x/y/z$ = 0.04/0.025/0 exhibited saturation polarization under an applied electric field of 50–60 kV/cm at 50 Hz with high P_r and low E_c. The value of polarization was observed to decrease with increasing frequency. This may be due to the fact that the contributions of all type of polarization are present at low frequency. As the frequency is increased, this type of contribution is decreased. The variations of P_r and E_c for all the samples were listed in Table 1.

The piezoelectric measurements of the poled BLNLT-BT system, sintered at 1150°C, for 2 hour, measured at room temperature were included in Table 1. With Ba^{2+} substitution, the piezoelectricity was reduced. According to thermodynamic theory of ferroelectricity, piezoelectric charge constant (d_{33}) is greatly dependent on the relative permittivity and polarization, which is directly proportional to dielectric permittivity (ε_r) and remnant polarization (P_r), as per the following equation [33]:

$$d_{33} = 2\varepsilon_r\varepsilon_0 Q_{11} P_r, \quad (2)$$

where ε_0, ε_r, and Q_{11}, are the vacuum/relative permittivity and electrostrictive coefficient, respectively, where Q_{11} should not change significantly by the doping for perovskite ceramics. Thus, value of d_{33} had been found to decrease due to weak ferroelectricity in BLNLT-BT system. Electrical properties of BLNLT-BT system had been compared with

Table 2: Dielectric and piezoelectric properties of [1 − z BLNLT-zBT] compared with the literature values.

	$z = 0$	$z = 0.02$	$z = 0.04$	$z = 0.06$	References in the literatures
ε_r(RT) /$\varepsilon_r(T_m)$	1365/3976	1386/3656	1423/3288	1494/4025	1080/3686 at T_m (NBT − BT$_{x/x=0.05}$) [34], 1164 [35], 924 (NBT − BT$_{x/x=0.11}$) [36], 863–1547/1 kHz (1 − xBLNT −xBT$_{x0.02-0.06}$) [37]
Tan δ(RT)	0.08	0.27	0.08	0.06	0.06 [34], 0.02 [35], 0.057 [36]
T_m (°C)	355	305	290	227	342 [34], 261 [36]
T_d (°C)	170	130	120	80	139 [34], 110 [36],
γ	1.69	1.58	1.66	1.79	1.9 [36]
D_{av} (μm)	2.67	2.34	—	1.78	2.5 [34]
D_{33} (pC/N)	70	65	47	20	77 [34], 118 (x = 0.04), 142 (x = 0.06) [38] 26 (x = 0.08) [35], 13 [36]

reported results (shown in Table 2). There were few reports, which are available on grain size, in which grain size played an important role to affect piezoelectric properties. The piezoelectric properties were expected to degrade with smaller grain size and improved the dielectric strength and mechanical strength [39], which may be explained on the basis of several models including the presence of internal stress in fine-grained ceramics, which was due to the absence of 90° domain walls, increased domain-wall contributions to the dielectric response in fine-grained ceramics, shift in phase transition temperatures with grain size, and so forth [33].

4. Conclusions

In summary, the complex BLNLT-BT system with compositions $x/y/z = 0.04/0.025/0$–0.06 was synthesized at temperature 1150°C by semiwet technique. The substitution of La/Li/Ba ($x/y/z$) had shown a significant effect on the microstructure, phase transition temperatures (T_d & T_m), dielectric, ferroelectric, and piezoelectric properties in BLNLT-BT system. Microstructure of all the specimens exhibited homogeneous grain growth, and the grain size was found to decrease with substitution of Ba in BLNLT-BT system. The temperature dependence of dielectric constant confirmed diffuse phase transition behaviour in BLNLT-BT system. There was a significant influence of the reduced grain size on the dielectric and piezoelectric properties of present system with Ba substitution.

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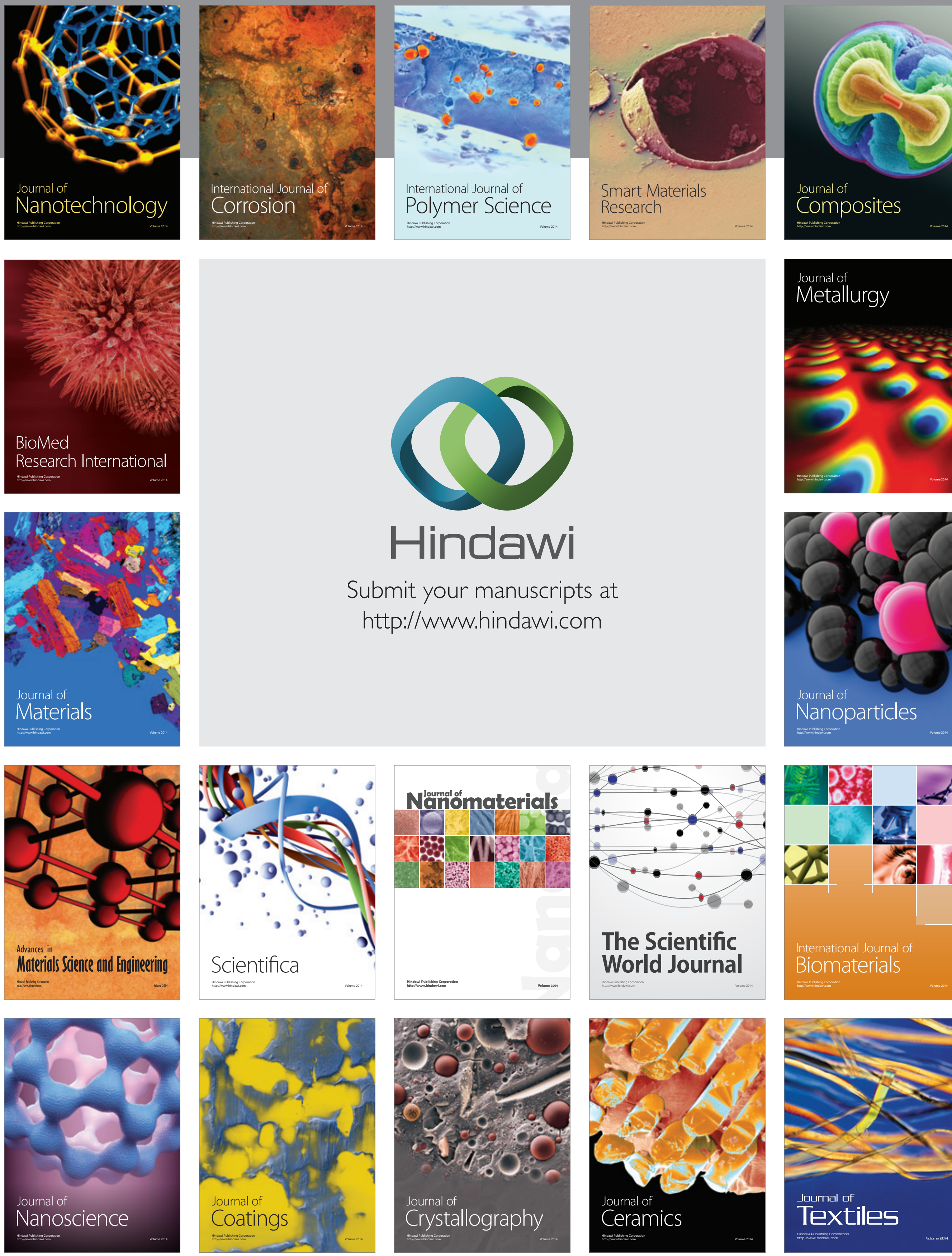
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