Journal of Electronic Materials (2022) 51:1068–1076
https://doi.org/10.1007/s11664-021-09351-3

ORIGINAL RESEARCH ARTICLE

Electrical Properties of Lead-Free $(Bi_{0.5}Na_{0.5})TiO_3$ Piezoelectric Ceramics Induced by BNT Nanoparticles

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Received: 23 July 2021 / Accepted: 15 November 2021 / Published online: 11 January 2022
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Abstract

$(Bi_{0.5}Na_{0.5})TiO_3$ (BNT) ceramics have been successfully synthesized by a seed-induced method using nanometric BNT particles as seeds. The effect of the BNT seeds on the phase formation, microstructure, and electrical properties was examined. The results show that all the ceramic samples had perovskite phase. The density values lay in the range from 5.87 g/cm^3 to 5.93 g/cm^3 with relative density values of 97.95% to -98.96%. The highest dielectric constant ε_r of 5138 at a maximum temperature of T_m was obtained for the seed sample with $x = 10$ mol.%. The highest piezoelectric charge coefficient (d_{33}) was 137 pC/N, and the piezoelectric voltage coefficient (g_{33}) was 20.72×10^{-3} Vm/N, being obtained for the seed samples with $x = 4$ mol.% and 8 mol.%, respectively. In addition, BNT seeds enhanced the energy storage density at room temperature. A high recoverable energy storage density W_{rec} of ~ 0.078 J/cm^3 was achieved for the sample with $x = 6$ mol.%. These results suggest that BNT seeds enhanced the electrical properties of the BNT ceramic.

Keywords BNT · lead-free ceramics · electrical properties · ferroelectric properties

Introduction

For many years, $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) has been reported to exhibit exceptionally high dielectric and piezoelectric properties. These ceramics are very important for many electronic applications.[1] However, PZT ceramics are not environmentally friendly due to the toxicity of lead oxide and its especially high vapor pressure during the sintering process at high temperatures. In recent years, many researchers have become interested in developing lead-free piezoelectric materials as alternatives to lead-based materials.[2-7] Sodium bismuth titanate ($Na_{0.5}Bi_{0.5}TiO_3$, NBT)[3,4,8-13] is one such lead-free ceramic that has been considered to be an important material for lead-free fabrication and a promising candidate for use in electronic applications because of its high Curie temperature (~ 340°C) and high remanent polarization (38 μC/cm^3). However, pure BNT ceramic has a large coercive field (E_c ~ 73 kV/cm)[8] and high conductivity that make the poling process difficult. Moreover, it has been found that data on the piezoelectric properties, especially d_{33} values, of pure BNT ceramic are scarce and show low values. To improve the electrical properties of lead-free BNT ceramics for applications, several processing methods have been used. Doping of rare-earth elements into BNT-based ceramics is one of the methods that is known to improve their electrical properties. Systems such as La-doped BNT,[14] Ce-doped BNT ($Bi_{0.5}Na_{0.44}K_{0.06}TiO_3$),[15] Er-doped BNT,[16] and Nd-doped BNT ($Bi_{1-x}Nd_xNa_{0.5}TiO_3$)[17] have been tested in the past. Also, many researchers have enhanced the electrical properties of BNT ceramics through the addition of other compounds such as $(Bi_{0.5}Na_{0.5})TiO_3$-$BaTiO_3$ (BNT-BT),[11,18,19] $(Bi_{0.5}Na_{0.5})TiO_3$-$Ba(Al_{0.5}Ta_{0.5})O_3$ (BNT-BAT),[20] and $Bi_{0.5}Na_{0.5}TiO_3$-$BaTiO_3$-$K_{0.5}Na_{0.5}NbO_3$ (BNT-BT-KNN).[21] Nevertheless, it is well known that the electrical properties of piezoelectric ceramics depend on not only their composition but also their texture. The crystallographic texture or grain orientations in the microstructure of the ceramics have been shown to have an effect. Methods to synthesize ceramic piezoelectric materials with texture to improve their electrical properties, e.g., to achieve high dielectric constant and excellent piezoelectric properties, have been reported, including the use of templated-grain growth[22-25] and seed-induced

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methods.[26-28] Those methods used nanoparticles as seeds or templates added to the base ceramic component. The addition of perovskite seeds may play an important role in the preparation of perovskite powders and films. The addition of seeds an help to induce the reactivity of the precursor, the homogeneity of the solid solution, and the formation of perovskite phase by decreasing the energy barrier and also reducing the calcination temperatures and dwell times. Moreover, the use of seeds may improve the electrical properties of piezoelectric ceramics.[26-28] Recently, Bai et al.[24] reported that the inclusion of plate-like $SrTiO_3$ could improve the piezoelectric coefficient (d^*_{33}) of $0.72(Bi_{0.5}Na_{0.5})TiO_3$-$0.28SrTiO_3$ (BNT-ST) ceramics. The textured ceramics showed enhanced dielectric properties and excellent piezoelectric coefficients with a strain level of 0.39% at 70 kV/cm and strain d^*_{33} (S_{max}/E_{max}) of 557 pm/V compared with the random ceramic samples. Moreover, <001>-oriented BNT-KN-ST ceramics with composition $0.90(Bi_{0.5}Na_{0.5})TiO_3$-$0.05KNbO_3$-$0.05SrTiO_3$ prepared by template-grain growth and using perovskite plate-like $(Bi_{0.5}Na_{0.5})TiO_3$ and $SrTiO_3$ particles as a template were reported by Bai et al.[25] The results of that study illustrated that the strain response increased with increasing texture. A large strain of 0.38% and normalized strain S_{max}/E_{max} of 540 pm/V were obtained in the textured samples obtained with the BNT template. Temperature stability over the range from room temperature to 120°C was found for the textured ceramic obtained using the BNT template.

In previous by work by our research team,[28] the effect of nanometric seeds on the phase formation and electrical properties of $Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$ (BCZT) ceramics prepared by the seed-induced method with different seed contents (0.0 mol.% to 4.0 mol.%) was studied. The results found that pure perovskite phase formed at low calcination temperatures. Some electrical properties, especially the d_{33} value, were enhanced with BCZT seeding. The samples with 3.0 mol.% seeds showed the highest phase-transition temperature (T_c) of ~93°C, the highest d_{33} value of ~ 520 pC/N, and the highest k_t value of ~48%.

As mentioned above, the seeding process or template grain growth (TGG) method can enhance the electrical properties of piezoelectric ceramics. Thus, in this work, the improvement of the structure and electrical properties of BNT ceramics prepared by the seed-induced method using BNT nanoparticles as seeds, and the effects of the seed content on the structure and electrical properties, were investigated.

Experimental Procedures

Lead-free $Bi_{0.5}Na_{0.5}TiO_3$ ceramics were produced by the seed-induced method using $Bi_{0.5}Na_{0.5}TiO_3$ nanoparticles as seeds. The seeds were synthesized from the starting materials of the $Bi_{0.5}Na_{0.5}TiO_3$ component by the molten-salt method. Starting powders of Bi_2O_3 (Sigma-Aldrich, ≥99.9%), $NaCO_3$ (Sigma-Aldrich, 99.95-100.5%), and TiO_2 (Sigma-Aldrich, 99-105.5%) were weighed and mixed with KCl-NaCl salt in ratio of 1:1, and the mixed powder was heated at 700°C for 2 h. After that, they were washed several times with hot deionized water until no trace of anion (Cl^-) was detected by using $AgNO_3$ solution. The powder was then dried in an oven at 120°C. The BNT seeds (BNTs) were then mixed with the raw materials of Bi_2O_3, $NaCO_3$, and TiO_2 at a ratio of BNT:seed = $(1 - x)$:x, where the BNT seed content was varied from 0 mol.% to 10 mol.%, being prepared by a solid-state reaction method. The mixed powder was ball-milled for 24 h and calcined at 900°C for 2 h. The powder was then mixed with an organic binder, pressed into pellets, and sintered at 1100°C for 2 h.

The phase formation and microstructure of the bulk ceramic systems were analyzed by x-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. To study the electrical properties, ceramic samples were polished and coated with silver paste to form electrodes. The dielectric constant, dielectric loss, and alternating-current (AC) conductivity of the sintered ceramics were measured as functions of frequency and temperature using an automatic dielectric measurement system (4980A, precision LCR meter; Agilent Technologies). To measure the ferroelectric properties, an electric field of up to 1 kV/mm to 7 kV/mm was applied to electrode-coated samples using a Sawyer-Tower circuit (Radiant Technology Inc.). Ferroelectric parameters such as the P-E loop, remanent polarization (P_r), and coercive field (E_c) were measured as functions of the BNT seed content. For the piezoelectric properties, the coated ceramics were poled at room temperature by applying an electric field of 6 kV/mm for 30 min in a silicone oil bath. After 24 h, the piezoelectric coefficient was measured using an S5865 d_{33} meter (KCF Technologies).

Results and Discussion

Lead-free $Bi_{0.5}Na_{0.5}TiO_3$ ceramic was successfully synthesized by using the same composition as seeds. $Bi_{0.5}Na_{0.5}TiO_3$ particles (BNTs) were successfully synthesized using the molten-salt method. The XRD pattern and microstructure are presented in Fig. 1. The results show that the phase structure of the BNTs calcined at 700°C had perovskite phase with a small amount of unknown phase. The BNTs showed a small particle sizes, as shown in the inset of Fig. 1. The XRD pattern was shown to be in close relation to the tabulated data of $Bi_{0.5}Na_{0.5}TiO_3$in JC file no. 00-046-0001 (JC file standard). The BNT was then mixed with BNT nanoparticle seeds (BNT-xBNTs) and sintered at 1100°C for 2 h. It was found that perovskite phase was obtained in the investigated composition range with a small amount of unknown impurity phase (Fig. 2). From Fig. 2a, the XRD patterns showed peaks corresponding to perovskite peaks of (100), (110), (111), (200), (210), and (211) (JC file standard no. 00-036-0340). The presence of perovskite phase in the ceramic system confirmed that the seed nanoparticles were successfully incorporated into the BNT lattice. The results indicate that the seed nanoparticles acted as a template for stable BNT perovskite formation.[26,27] The expanded XRD patterns in the range of 40° to 48° are shown in Fig. 2b.

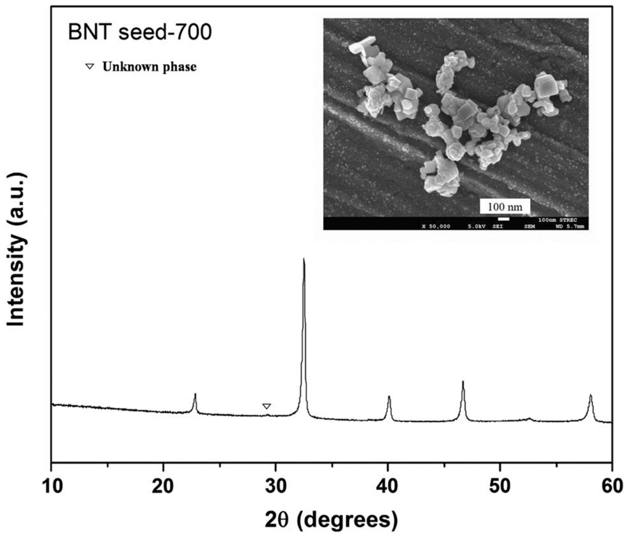

Fig. 1 XRD pattern and SEM micrograph for BNT seed nanoparticles.

It was found that the XRD peaks of all the samples corresponded to rhombohedral (R) state, as observed by the splitting peaks of (111)/($1\bar{1}1$) reflections at $2\theta \sim 40°$ to 42° and single peak of (200) reflection at $2\theta \sim 46°$ to 48°.[29-31] The bulk density, relative density, and grain size values of the ceramic samples are presented in Table I. Figure 3 shows SEM micrographs and the distribution percentage of sintered surface for the BNT-BNTs ceramics. The density and grain size were measured using the Archimedes principle and the line intercept method, respectively. It can be seen that the BNT seeds clearly affected the density and grain size of the ceramic. The results show that grains of the seed-free sample (0BNTs) showed a square shape but changed to a polygonal shape for the sample with 2 mol.% to 8 mol.% seeds added. Again, the sample with 10 mol.% seeds (10BNTs) clearly showed a square shape with large grains. Moreover, the surface of the ceramic samples with 2 mol.% to 8 mol.% seeds added showed uniform grains and homogeneous microstructure with no large pores. As seen in Fig. 3, the grains were well faceted, in good accordance with the high crystallinity of the fabricated samples (Figs. 1, 2). It can be suggested that the relationship between the faceted microcrystal shapes and high crystallinity is general for solids, being observed for different compounds.[32,33] A highly homogeneous microstructure distribution was observed in the seeded samples, which may be a result of the nanometric seed particles' acting as nuclei for homogeneous grain growth in the BNT ceramics.[26]The grain size of the ceramics was in the range of 1.41 μm to 2.95 μm. In the case of the

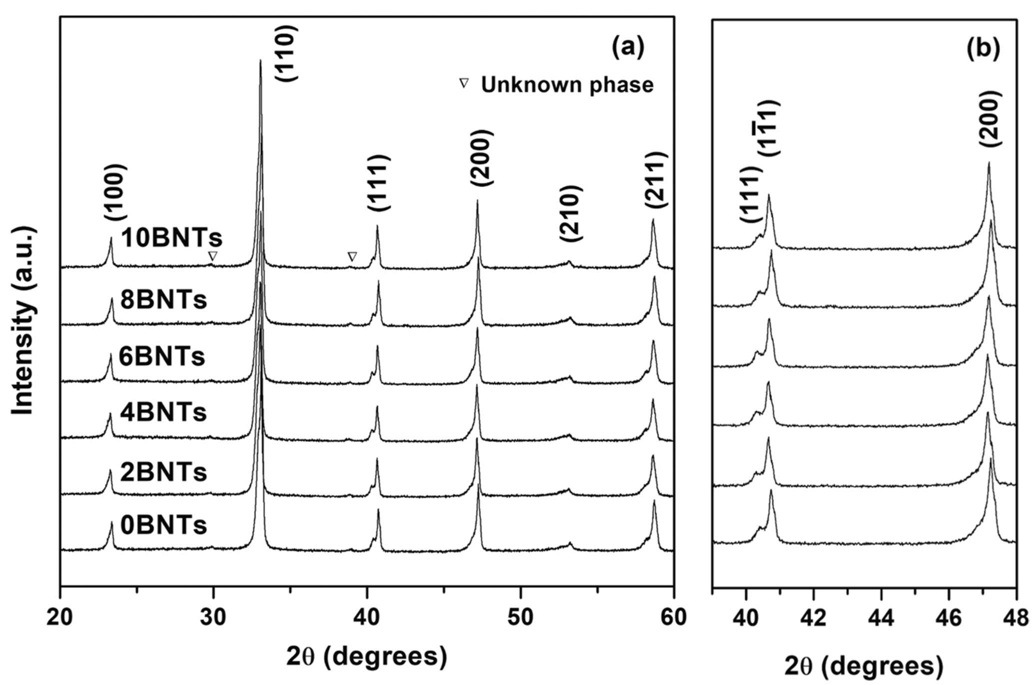

Fig. 2 (a) XRD patterns and (b) expanded XRD patterns for the 2θ range of 40° to 48° for sintered BNT-BNTs ceramic samples.

Table I Density, relative density, and dielectric properties of all the ceramic samples

Sample	Density (g/cm^3)	Relative Density (%)	Grain Size (μm)	Dielectric ε_r At T_r	Loss tan δ	Dielectric ε_r At T_m	Loss tan δ	T_m (°C)
0	5.87 ± 0.007	97.95	1.94	793.04	0.0469	4467.50	0.2309	350
2	5.93 ± 0.006	98.96	1.41	757.07	0.0467	4437.52	0.3387	349
4	5.91 ± 0.003	98.58	1.46	762.02	0.0450	4228.44	0.1936	339
6	5.90 ± 0.016	98.46	1.60	738.51	0.0469	3806.03	0.2268	345
8	5.89 ± 0.012	98.38	1.48	733.74	0.0518	3793.15	0.2685	340
10	5.88 ± 0.006	98.14	2.95	769.02	0.0441	5138.50	0.6025	357

T_r is room temperature (data obtained at a frequency of 1 kHz)

T_m is maximum temperature (data obtained at a frequency of 1 kHz)

samples with 2 mol.% to 8 mol.% seeds added, the grain size tended to increase with increasing BNT seed content from 2 mol.% to 6 mol.% but then decrease for the sample with 8 mol.% BNTs. It is evident from the data presented in Table I that the samples with added seeds showed higher density values than the seed-free sample. These findings may be due to the seed nanoparticles increasing the diffusion flux and the densification rate for the ceramics. It is well known that the sintering process, in terms of the lattice diffusion of vacancies from pores to grain boundaries, leads to densification.[34] Furthermore, the higher degree of densification may be related to the uniform grain and homogeneous grain size distribution of the ceramic systems.[26] In this study, the density increased from 5.87 g/cm^3 for the seed-free sample (BNTs = 0.0) to 5.93 g/cm^3 for the sample with x = 2.0 mol.% added seeds. It then tended to decrease with further increase in the seed content. The density values were shown to lie in the range from 5.87 g/cm^3 to 5.93 g/cm^3 with a theoretical density of 97.95% to 98.96%.

The dielectric constant (ε_r) and dielectric loss (tan δ) measured at room temperature and 1 kHz are presented in Table I. The results reveal that the dielectric constant (ε_r) tended to decrease with an increase in the seed content. This decrease in the dielectric constant may have been the result of structural disorder, which is a consequence of different valences, ionic radii, and oxygen vacancies. It is well known that the dielectric constant is dependent on the crystal structure and microstructure.[35] The decreasing dielectric constant in the present study may be due to a change of the crystal structure. The dielectric loss (tan δ) showed no significant variation with the BNT seed content. The dielectric constant and dielectric loss were in the range of 733.74 to 793.04 and 0.0441 to 0.0518, respectively. Figure 4 shows the dielectric constant (ε_r) and dielectric loss (tan δ) as functions of temperatures in the range from 30°C to 450°C and of frequency in the range from 1 kHz to 100 kHz. Plots of ε_r measured at room temperature and the temperature with the maximum ε_r (at 1 kHz) with x mol.% seeds (xBNTs) are shown in Fig. 5 while the data are presented in Table I.

In relation to Fig. 4, it was found that the ε_r curves showed broad peaks and dispersion with frequency for all conditions. This suggests that the ceramics exhibited a relaxor ferroelectric state.[36] In this study, the dielectric curves showed two dielectric anomalies for all ceramic samples: a shoulder at low (~ 200°C) and a maximum at high (~350°C) temperature, corresponding to known transitions from rhombohedral phase to antiferroelectric (tetragonal phase) state and antiferroelectric (tetragonal) to paraelectric (tetragonal) phase, respectively.[18,37,38] In addition, note that the ceramic systems exhibited coexistence of the rhombohedral and tetragonal phases with polar regions.[39] T_m did not change significantly with the seed content in the range from $x = 0$ mol.% to $x =8$ mol.%. The maximum value of ε_r tended to decrease from 4467 for the seed-free sample ($x = 0$ mol.%) to 3793 for the sample with $x = 8$ mol.% seeds added and then to increase to 5138 for the sample with x = 10 mol.% BNT seeds (Fig. 5; Table I). Based on the results presented in Table I, the tan δ values showed little change with increasing seed content. However, the tan δ value for the sample with 10 mol.% seeds was the highest, which may be because of its highest dielectric constant.

The AC conductivity (σ_{AC}) of the BNT-BNTs ceramics measured at room temperature is shown in Fig. 6. The AC conductivity (σ_{AC}) can be defined by Eq. 1:[40]

$$\sigma_{AC} = \omega\varepsilon_0\varepsilon_r \tan\delta, \tag{1}$$

where ω is the angular frequency ($\omega = 2\pi f$), f is frequency, ε_0 and ε_r are the permittivity of vacuum and the dielectric constant, and tan δ is the dielectric loss. As seen in Fig. 6, the AC conductivity was strongly dependent on frequency, as observed from the straight-line slope of the σ_{AC} plots. The σ_{AC} values tended to increase with frequency for all the samples. From Fig. 6, it can be seen that the σ_{AC} values decreased when the BNT seed content was increased from x = 2 mol.% to x = 6 mol.% then increased with further increase of the BNT seed content. Note that the increasing σ_{AC} value corresponds to a decreasing resistance. This

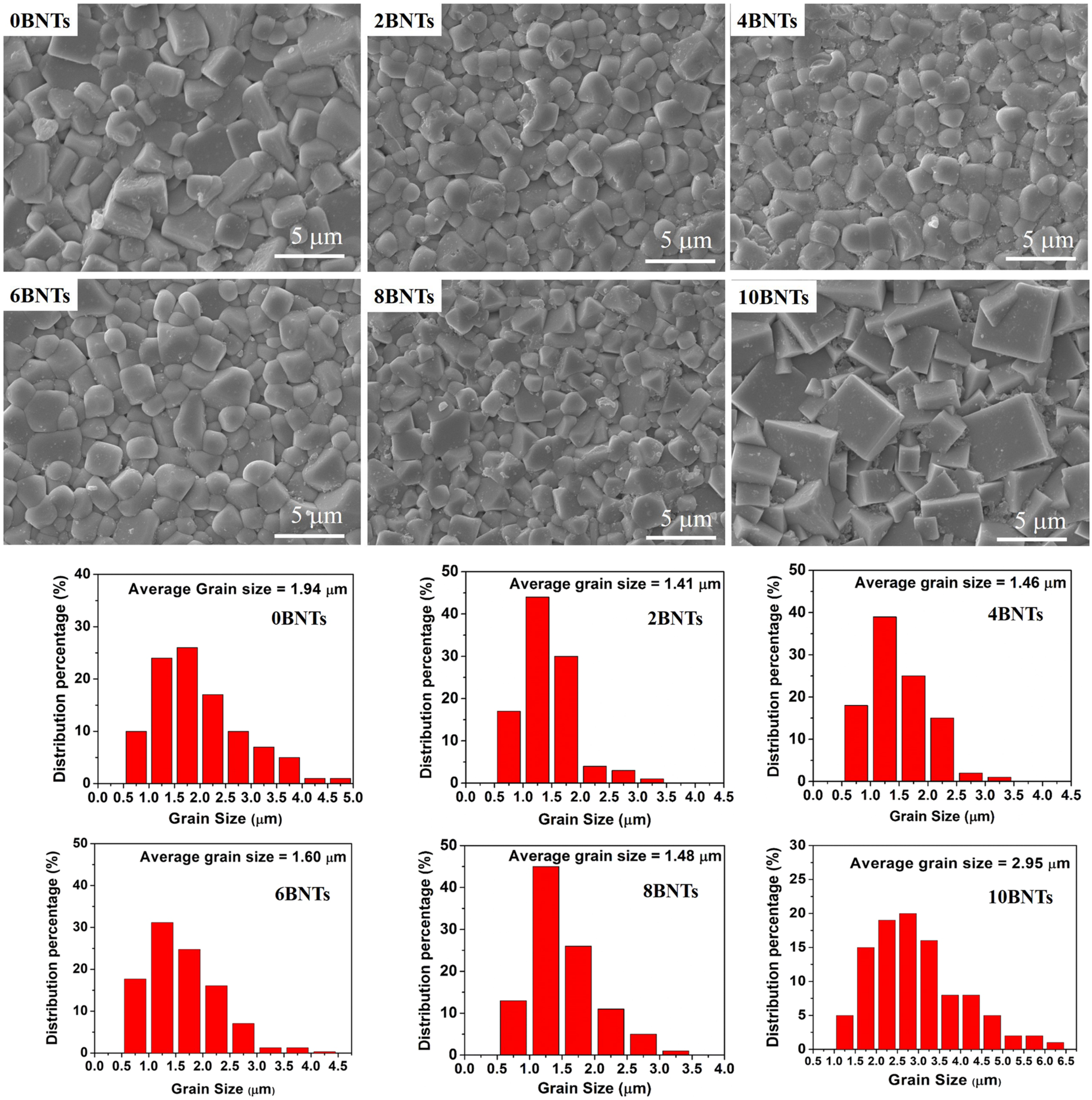

Fig. 3 SEM micrographs and distribution percentage of sintered BNT-BNTs ceramics.

suggests that the BNT seeds increased the resistance of the BNT ceramic. Moreover, the dependence of the AC conductivity on frequency is a mechanism for the low-temperature region which results from charge carriers and space-charge polarization in the structure or extrinsic dipoles of impurities.[40,41] These results could be caused by impurities in each ceramic condition.

The relations between the polarization and the electric field (P-E hysteresis loop) measured at room temperature are shown in Fig. 7a. The plots of the ferroelectric parameters, including the remanent polarization (P_r) and coercive field (E_c) as functions of the BNT seed content are shown in Fig. 7b and presented in Table II. It can be seen that all the ceramics showed ferroelectric behavior with a large loop, and the P-E loops for the samples with added seeds changed in comparison with that of the seed-free sample. The seeds thus affected the remanent polarization (P_r) and coercive field (E_c) of the BNT ceramic. First, the P_r value dropped in the sample with a seed content of 2 mol.%, then tended to increase with further increase in the

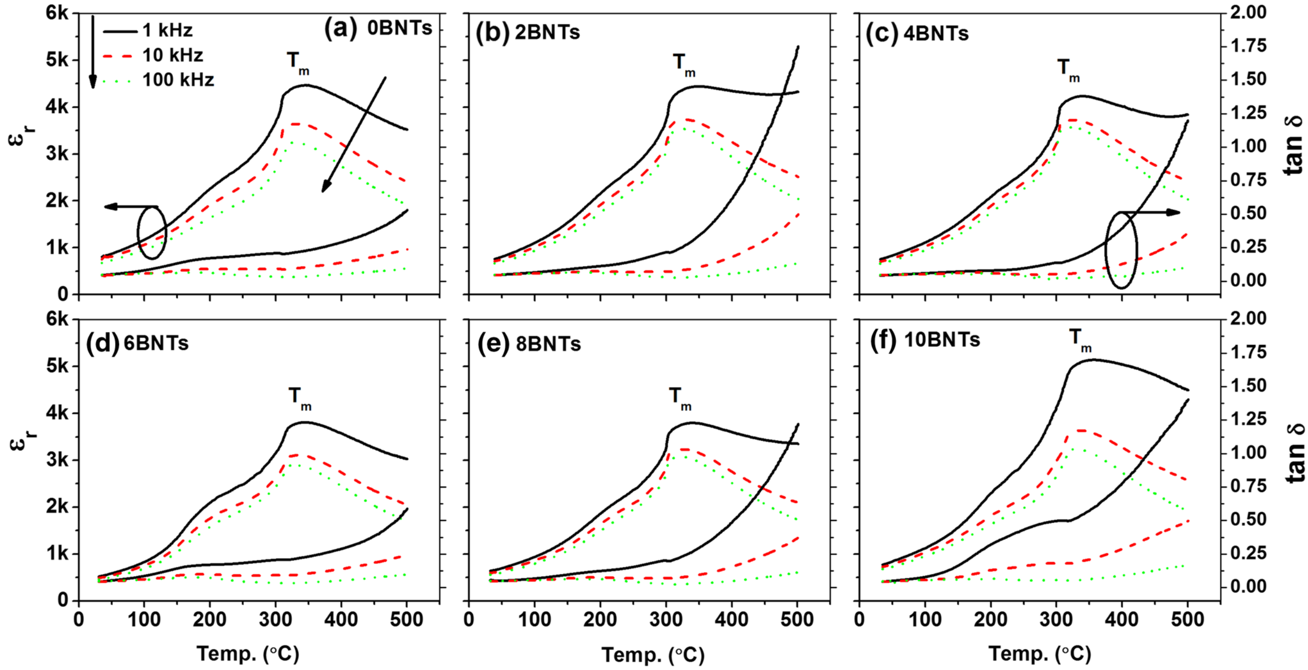

Fig. 4 Dielectric constant (ε_r) and dielectric loss (tan δ) as functions of temperature for the BNT-BNTs ceramics.

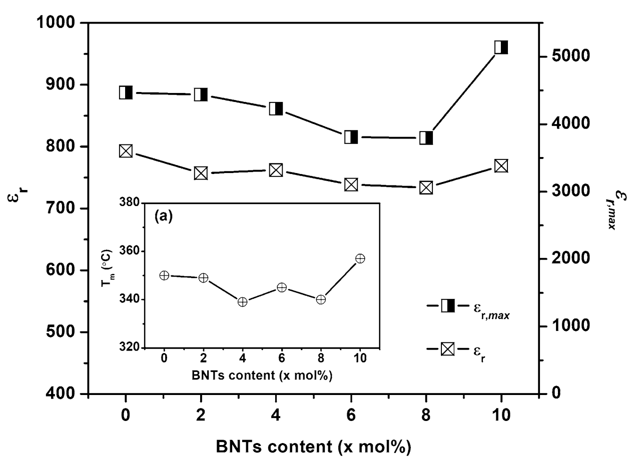

Fig. 5 Dielectric constant at room temperature and maximum temperature (T_m, inset) for various BNT-BNTs ceramics.

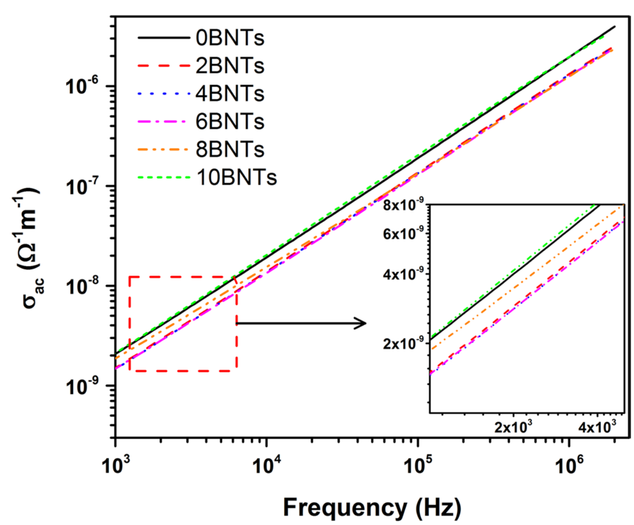

Fig. 6 Ac conductivity (σ_{AC}) of BNT-BNTs ceramics.

seed content to 4 mol.%, 6 mol.%, and 8 mol.%, but then decreased for the sample with 10 mol.% seed content. It is well known that ferroelectric properties are affected by the composition, uniform oriented domains, electric field, defects, and grain size.[42] The mentioned decrease of P_r may result from modification of the structure with addition of the BNT seeds. These results are similar to previous findings by Bai et al.,[25] who reported on the effect of a BNT template on the piezoelectric properties of BNT-base ceramics. In addition, the P_r value drop observed for the sample with 2 mol.% seeds may be due to the decreasing grain size. The E_c value showed less change as the seed content was varied from 2 mol.% to 8 mol.%. The P_r and E_c values of the ceramics lay in the range from 26.32 $\mu C/cm^3$ to 38.40 $\mu C/cm^3$ and 54.54 kV/cm to 55.57 kV/cm, respectively.

The recoverable energy storage density (W_{rec}) of the ceramics was calculated by integrating the area of the *P-E* hysteresis loops according to Eq. 2;[43,44]

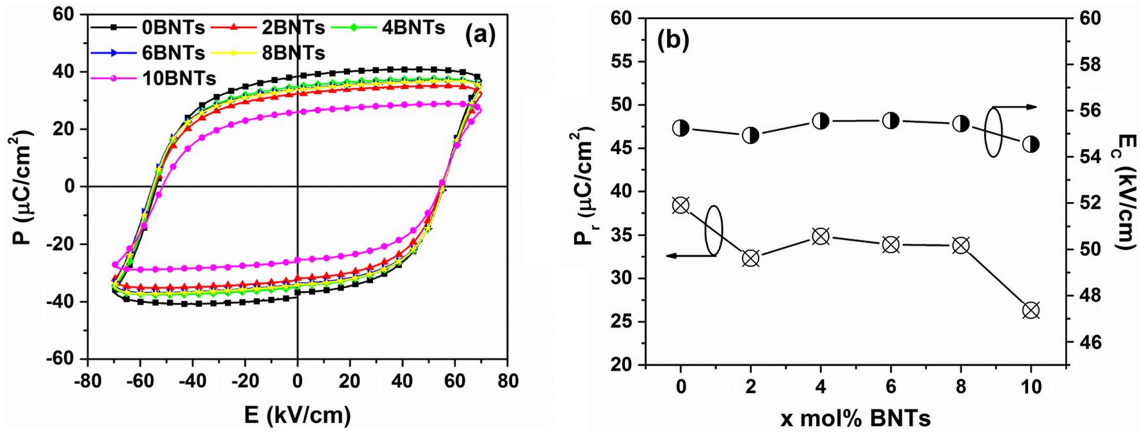

Fig. 7 *P-E* hysteresis loops (a) and ferroelectric parameter; remanent polarization (P_r) and coercive field (E_c) (b) of BNT-BNTs ceramics measured at room temperature

Table II Ferroelectric parameters and piezoelectric coefficient for BNT-BNTs ceramics

Sample	P_r (μC/cm^3)	E_c (kV/cm)	W_{rec} (J/cm^3)	d_{33} (pC/N)	g_{33} ($\times10^{-3}$ Vm/N)
0	38.40	55.24	0.045	114.2	16.26
2	32.31	55.00	0.069	116.8	17.42
4	34.85	55.55	0.062	136.6	20.25
6	33.90	55.57	0.078	130.4	19.94
8	33.80	55.44	0.076	134.6	20.72
10	26.32	54.54	0.064	127.0	18.65

$$W = \int_{P_r}^{P_{max}} E\,dp, \tag{2}$$

where W is the energy storage density, E is the applied external electric field, P_{max} is the maximum polarization, and P_r is the remanent polarization.

To obtain a high recoverable energy storage density (W_{rec}), a large dielectric breakdown strength (DBS), reduced remanent polarization (P_r), and increased maximum polarization (P_{max}) are required.[43,45,46] In this work, the W_{rec} value of the ceramics is presented in Table II. It was shown that, for an applied electric field of 70 kV/cm (measured at room temperature), the W_{rec} value of the samples with added seeds was higher than that of the seed-free sample and reached a maximum value of 0.078 J/cm^3 for the sample with 6 mol.% BNT seeds. The higher W_{rec} value for the samples with added seeds may have resulted from the lower remanent polarization (P_r).

The piezoelectric charge coefficient (d_{33}) and piezoelectric voltage coefficient (g_{33}) of the BNT-BNTs ceramics are presented in Table II. It can be seen that the BNT ceramics with added BNT seeds exhibited higher values of d_{33} and g_{33} than the pure BNT sample. The d_{33} value clearly increased from 114 pC/N for the pure BNT to around 137 pC/N for the samples with 4 mol.% to 8 mol.% seeds. The highest d_{33} value was obtained for the sample with 4 mol.% seeds. The g_{33} value showed the same trend as the d_{33} value for all the samples. The g_{33} values were determined by using the expression[47]

$$g_{33} = \frac{d_{33}}{\varepsilon_r \varepsilon_0}, \tag{3}$$

where ε_o is the permittivity of free space and ε_r is the relative permittivity. The values of ε_r were measured at room temperature. The g_{33} values lay in the range from 16.26×10^{-3} Vm/N to 20.72×10^{-3} Vm/N. The highest g_{33} value was obtained for the sample with $x = 8$ mol.%. From the d_{33} and g_{33} results, it can be suggested that the BNT nanoparticles enhanced the piezoelectric properties of the BNT ceramic. The higher d_{33} and g_{33} values for the samples with added seeds may be related to the increased density of the ceramics. However, the d_{33} value for the sample with 2 mol.% seeds was lower than those for the other samples with added seeds. This may be because of its smaller grain size. The piezoelectric coefficient d_{33} in this work was enhanced after addition of seeds. The results of this study are similar to the findings of Bai et al.[25] and Ye et al.,[23] respectively, when working on BNT-based ceramics and BCZT-based ceramics with the use of a template.

Conclusions

A seed-induced method was applied to synthesize lead-free BNT ceramic samples using BNT particles as seeds. The results showed average density values in the range from 5.87 g/cm^3 to 5.93 g/cm^3 with relative density values of 97.95% to 98.96%. Piezoelectric coefficient d_{33} values from 130 pC/N to 137 pC/N and g_{33} values from 19.94×10^{-3} Vm/N to 20.72×10^{-3} Vm/N were obtained for the composition range of $x = 4$ mol% to 8 mol.% seeds. A high recoverable energy storage density W_{rec} of ~ 0.078 J/cm^3 was achieved for the sample with $x = 6$ mol.%. These results confirm that addition of BNT seeds can improve the electrical properties of BNT ceramics.

Acknowledgments The authors thank the Thailand Research Fund (TRF) under the Research Grant for new scholar (MRG6180111), Faculty of Science and Technology and Rajamangala University of Technology Krungthep under the Research and Development Institute for financial support.

Conflict of interest The authors declare that they have no conflicts of interest

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