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Comparative Study of Structural, Microstructural, Dielectric, and Energy Storage Properties of $Bi_{0.5}Na_{0.5}TiO_3$ and $SrTiO_3$-doped $Bi_{0.5}Na_{0.5}TiO_3$ Ceramic

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Abstract

In this study, we investigated the outcome of $SrTiO_3$ (ST) doping on the performance of energy storage of $Bi_{0.5}Na_{0.5}TiO_3$ (BNT)-based ceramics. A 0.8BNT–0.2ST composition was synthesized via the conventional solid-state route and characterized for structural, microstructural, and dielectric properties. X-ray diffraction confirmed the formation of a single-phase P4bm perovskite structure, and FESEM analyses showed a noticeable reduction in grain size with 20wt% ST doping. A reduction in the dielectric constant was observed, along with a downward shift in the phase transition temperature. Consequently, an obvious improvement in recoverable energy density (W_{rec}) and efficiency (η) had been noticed from ferroelectric results, reflecting enhanced relaxor characteristics in the doped specimen. Overall, $SrTiO_3$ addition was found to be an effective strategy for boosting the energy storage capability of BNT ceramics.

1.Introduction

As the demand for efficient and reliable energy storage solutions grows, ferroelectric ceramics have gained attention because of their high energy density, fast charge and discharge capabilities, and eco-friendly characteristics [1,2]. $Bi_{0.5}Na_{0.5}TiO_3$ (BNT) is notable as a lead-free ferroelectric material with promising capabilities. However, its practical use is limited due to its large remanent polarization and comparatively low energy storage efficiency [3]. To overcome these issues, researchers have widely explored doping as a way to improve both the structure and electrical properties. Adding $SrTiO_3$ (ST), a paraelectric perovskite, into the BNT framework can disrupt the long-range ferroelectric ordering, promote relaxor-like characteristics, and enhance the material's energy storage performance [4]. In this work, BNT and 0.8BNT–0.2ST ceramics were prepared and thoroughly analyzed to investigate their structural, dielectric, and ferroelectric properties. Special attention was given to the impact of adding ST on phase composition, grain structure, dielectric behavior, and polarization–electric field (P–E) loops, which are important for improving capacitive energy storage performance.

2. Materials and Experimental Methods

A conventional solid-state reaction method was used to synthesize $Bi_{0.5}Na_{0.5}TiO_3$ (BNT) and $0.8Bi_{0.5}Na_{0.5}TiO_3$–$0.2SrTiO_3$ (0.8BNT–0.2ST) ceramics. Highly pure precursors—Na_2CO_3, Bi_2O_3, $SrCO_3$, and TiO_2 (all >99.9%, Hi-Media)—were weighed according to stoichiometry and ball-milled in ethanol for 24 hours. After drying and calcining the slurry at 100 °C and 850 °C, the powders were subsequently ball-milled for an additional 10 hours. Polyvinyl alcohol (PVA) served as the binder before uniaxial pressing into pellets (diameter of 12 mm, ~1 mm thickness), followed by sintering at 1100 °C. X-ray diffraction (XRD) pattern results were attained with the help of a Rigaku Smart Lab diffractometer with Cu-Kα radiation (λ = 1.5406 Å), scanning from 20° to 80° (2θ) at 0.02° step size and 2°/min. Microstructural studies were conducted utilizing field emission scanning electron microscopy (FESEM, Carl-Zeiss-Ultra Plus) on thermally etched surfaces. Grain size distributions were analyzed using ImageJ software. For electrical measurements, silver paste was applied to polished pellets and fired at 250 °C for 30 min. Temperature- and frequency-dependent dielectric properties were obtained utilizing an HIOKI 3532-50 LCR Hi-Tester. Ferroelectric polarization-electric field (P–E) loops were recorded in silicone oil at 50 Hz using a ferroelectric loop tracer (Marine India). Energy storage parameters such as Wrec and η were derived from the P–E loops.

3. Results and Discussions

Figure 1(a) displays the XRD pattern results of undoped BNT and 0.8BNT–0.2ST ceramics. Both samples exhibit characteristic peaks of the perovskite structure and match well with the standard JCPDS card #96-210-2069 (space group: P4bm), indicating phase purity. No secondary phases were observed within the detection limit. Notably, a slight shift in peak position upon ST doping suggests minor lattice

distortion because of the incorporation of bigger Sr^{2+} ions in the A-site and Ti^{4+} in the B-site [5]. FESEM images in Fig. 1(b) and 1(c) reveal significant grain size refinement upon ST incorporation. The undoped BNT shows an average grain size of ~3.6 µm, while the doped 0.8BNT–0.2ST sample exhibits a much finer average grain size of ~0.91 µm. This grain size reduction is beneficial for enhancing breakdown strength and improving energy storage performance [6]. The relatively uniform grain distribution in the doped sample also suggests improved densification and grain growth control due to the presence of ST.

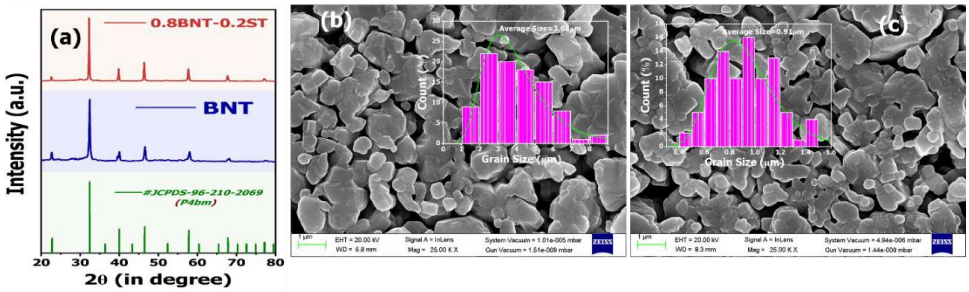

Figure 1: (a)XRD diffraction patterns and corresponding peak position; (b, c) FESEM micrographs and grain size distribution curves of BNT and 0.8BNT-0.2ST, respectively

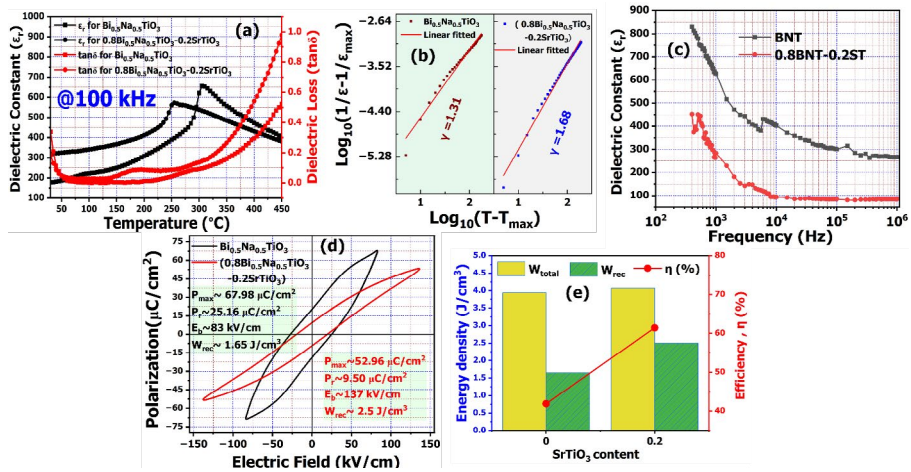

Figure 2: (a) Temperature-dependent ε_r and tan δ at 100 kHz for both compositions; (b) Modified Curie–Weiss fit showing γ values indicating relaxor behavior; (c) Frequency-dependent ε_r showing reduced dielectric constant and dispersion in doped sample; (d) P–E loops showing enhanced energy storage properties in 0.8BNT–0.2ST; (e) Comparison of energy density and efficiency for BNT and 0.8BNT–0.2ST

Temperature-dependent dielectric measurements at 100 kHz (Fig. 2(a)) show broad dielectric peaks after doping, corresponding to the maximum permittivity temperature (T_{max}), indicative of relaxor-type behavior. ST doping significantly reduces ε^r but broadens the permittivity peak, indicating enhanced relaxor behavior. The dielectric loss (tan δ) remains below 0.1 up to 300 °C for the doped sample, confirming good thermal stability. To quantitatively analyze relaxor behavior, the modified Curie–Weiss law was applied [7,8].

$$\frac{1}{\varepsilon_r} - \frac{1}{\varepsilon_{max}} = \frac{(T-T_{max})^{\gamma}}{C}$$ The calculated γ values (Fig. 2(b)) are 1.31 for BNT and 1.68 for 0.8BNT–0.2ST, indicating a normal to strong relaxor ferroelectric transition behavior due to ST doping. Frequency-dependent dielectric measurements (Fig. 2(c)) show a decreasing trend in ε_r with frequency, more pronounced in doped samples, which is typical for relaxor ferroelectrics [9]. The lower dielectric constant at high frequencies for 0.8BNT–0.2ST can be attributed to suppressed domain wall contributions and enhanced polar nano-regions (PNRs) stability.

Figure 2(d) displays the P–E loops for both compositions. The undoped BNT exhibits a typical ferroelectric loop with high remnant polarization ($P_r \approx 25.16\ \mu C/cm^2$) and maximum polarization ($P_{max} \approx 67.98\ \mu C/cm^2$). In contrast, the 0.8BNT–0.2ST sample exhibits slimmer and more symmetric loops with significantly reduced P_r (≈ 9.50 µC/cm²) and Pma_x (≈ 52.96 µC/cm²), which is advantageous for applications in energy storage. The W_{rec} and η were derived using: $W_{tot}=\int_0^{P_{max}} EdP$; $W_{rec}=\int_{P_r}^{P_{max}} EdP$; and $\eta=\frac{W_{rec}}{W_{tot}}$. As shown in Fig. 2(e), W_{rec} increases from 1.65 to 2.51 J/cm³, while η improves from 41.5% to 57.6% upon ST doping. The enhancement in energy storage properties is primarily due to reduced P_r, improved breakdown strength ($E_b \approx 137$ kV/cm), and induced relaxor behavior. These results demonstrate that ST doping effectively tunes the structure and polarization response of BNT ceramics, making 0.8BNT–0.2ST a promising candidate

for energy storage capacitors.

4. Conclusion

$Bi_{0.5}Na_{0.5}TiO_3$ and $0.8Bi_{0.5}Na_{0.5}TiO_3$–$0.2SrTiO_3$ dielectric ceramics were successfully synthesized using the solid-state method. ST doping led to refined microstructure, induced relaxor ferroelectric behavior, and enhanced dielectric stability. The doped sample exhibited a slimmer P–E loop with reduced remnant polarization and improved energy storage performance. The maximum recoverable W_{rec} of 2.51 J/cm^3 and η of 61.42% were attained for 0.8BNT–0.2ST, compared to 1.65 J/cm^3 and 41.88% for BNT. These enhancements are attributed to the suppressed long-range ferroelectric ordering and reduced grain size. Thus, $SrTiO^3$ doping offers a viable strategy to progress the capacitive performance in energy storage of BNT-based lead-free ceramic dielectrics.

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