

EFFECTS OF Li_2CO_3 CONTENT ON THE SINTERING BEHAVIOR AND PHYSICAL PROPERTIES OF PSLZT-BMT-PMS CERAMICS

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Abstract. This study presents the successful synthesis of $0.970[(\text{Pb}_{0.94}\text{Sr}_{0.05}\text{La}_{0.01})(\text{Zr}_{0.51}\text{Ti}_{0.49})\text{O}_3]-0.024[\text{Bi}(\text{Mn}_{1/2}\text{Ti}_{1/2})\text{O}_3]-0.006[\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3]$ ceramics doped with x wt.% Li_2CO_3 ($x = 0.0-1.0$) using the conventional solid-state method. The incorporation of Li_2CO_3 effectively promotes densification through liquid-phase-assisted sintering, enabling a reduced sintering temperature of 1050°C while simultaneously tailoring the defect chemistry and domain structure of the ceramics. Herein, the formation of a single-phase perovskite crystal structure, uniform elemental distribution, and the presence of morphological phase boundaries (MPBs) in the range of $0.4-0.6$ wt.% Li_2CO_3 is confirmed through X-ray diffraction (XRD) and energy-dispersive spectroscopy (EDS) analyses. The corresponding hard piezoelectric ceramic at the optimal concentration ($x = 0.6$ wt.%) exhibits good dielectric properties, with a dielectric constant (ϵ) of 1196, a piezoelectric coefficient (d_{33}) of 430 pC/N, an electromechanical coupling coefficient (k_p) of 0.60, a mechanical quality factor (Q_m) of 1220, and a low dielectric loss ($\tan\delta$) of 0.005. The enhanced performance is attributed to the synergistic effects of improved densification, defect dipole formation, and MPB-induced phase instability. These findings demonstrate that Li_2CO_3 not only acts as a sintering aid but also plays a critical role in defect-mediated domain engineering, providing an effective strategy for optimizing high-performance hard piezoelectric ceramics for high-power ultrasonic and actuator applications.

Keywords: Sintering Behavior, Li_2CO_3 , PZT-based materials, PSLZT-BMT-PMS.

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1 Introduction

Lead zirconate titanate (PZT)-based piezoelectric materials, particularly modified systems such as PSLZT and complex perovskite solid solutions incorporating Bi-based compounds, are extensively used in sensors, actuators, ultrasonic transducers, and high-performance piezoelectric transformers due to their superior electromechanical properties and compositional flexibility (Liu et al., 2025; Siddiqui et al., 2017; Wu et al., 2017; Pu et al., 2022). In particular, multi-component systems combining soft and hard dopants have attracted significant attention because they enable the simultaneous optimization of piezoelectric activity and mechanical quality factor (Q_m), which is critical for high-power applications (Zou et al., 2026). However, conventional PZT-based ceramic systems often require high sintering temperatures ($\geq 1150^\circ\text{C}$), resulting in PbO evaporation, non-homogeneous microstructure, degradation of electrical properties, and increased production costs (Pu et al., 2022; Lin et al., 2023). Therefore, reducing the sintering temperature while maintaining or enhancing functional properties remains a key challenge in the development

of advanced piezoelectric ceramics. Numerous studies have focused on identifying solutions to reduce the sintering temperature while maintaining the material's physical properties, including the use of nano-sized starting materials (Ghasemifard et al., 2009), sintering aids (Gao et al., 2022; Bi et al., 2025), hot pressing (Cao et al., 2025), and plasma sintering (Niemiec et al., 2023). Among these approaches, the use of sintering aids is particularly common, as they typically have low melting points or combine with other components to form a liquid phase at temperatures below the sintering temperature, thereby promoting ceramic densification (Zhou et al., 2025). Several metal oxide sintering aids have been incorporated into the PZT composition to effectively reduce the sintering temperature without reducing the electromechanical properties of the material, such as Li_2CO_3 (Pu et al., 2022; Gao et al., 2022; Vuong, 2023; Zhang et al., 2025), Bi_2O_3 (Zhang et al., 2024). In particular, lithium carbonate (Li_2CO_3) is an effective sintering aid for complex PZT ceramic systems. With a low melting point of roughly 723°C , Li_2CO_3 readily combines with PbO to create the low-melting-point liquid phase Li_2PbO_3 (approximately 836°C), significantly improving material movement during sintering. High-density ceramics can be produced at much lower temperatures than in conventional solid-phase sintering, as this transient liquid phase facilitates grain rearrangement and densification (Lin et al., 2023; Donnelly et al., 2009). Researchers have reported the effectiveness of Li_2CO_3 in various PZT-based material systems. For instance, Siddiqui et al. (2017) demonstrated that the addition of 0.7 mol.% Li_2CO_3 to $\text{Pb}_{0.93}\text{La}_{0.02}\text{Sr}_{0.05}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ (PLSZT) ceramic reduced the sintering temperature from 1150 to 850°C while maintaining good electrical properties. These properties include $\rho = 7.421 \text{ g/cm}^3$, $k_p = 0.46$, $d_{33} = 259 \text{ (pC/N)}$, $\varepsilon_r = 1270$, and $\tan \delta = 0.0847$. According to Zhang et al. (2017a), the piezoelectric ceramics $0.08\text{Pb}(\text{Ni}_{1/3}\text{Nb}_{2/3})\text{O}_3 - 0.02\text{Pb}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3 - 0.90\text{Pb}(\text{Zr}_{0.5}\text{Ti}_{0.5})\text{O}_3 + 0.30 \text{ wt.\% } \text{CaCO}_3 + 0.2 \text{ wt.\% } \text{Li}_2\text{CO}_3$, sintered at 900°C for four hours, exhibited the following excellent electrical characteristics: $d_{33} = 599 \text{ pC/N}$, $k_p = 0.698$, $T_C = 296^\circ\text{C}$, $\varepsilon_r = 2595$ (1 kHz), $\tan \delta = 0.0174$ (1 kHz), $P_r = 38.25 \text{ } \mu\text{C/cm}^2$ (20 kHz), and $E_c = 10.03 \text{ kV/cm}$ (20 kHz). Furthermore, Zhang et al. (2022) reported the successful synthesis of $15\text{PSN} - 52\text{PMN} - 33\text{PT} + 0.5 \text{ wt.\% } \text{Li}_2\text{CO}_3$ piezoelectric ceramics at 1100°C using the two-step columbite precursor method. The resulting ceramic exhibited excellent piezoelectric properties, including $d_{33} = 520 \text{ pC/N}$, $k_p = 0.62$, $T_c = 180^\circ\text{C}$, $E_c = 5.1 \text{ kV/cm}$, and $P_r = 33.6 \text{ } \mu\text{C/cm}^2$. According to Quang & Vuong (2022), the PZT–PZN–PMnN ceramic system co-doped with 0.07 wt.% Li_2CO_3 and 0.25 wt.% Fe_2O_3 was successfully synthesized at 1000°C . The obtained ceramics exhibited excellent dielectric and piezoelectric performance, including $d_{33} = 455 \text{ pC/N}$, $Q_m = 1455$, and $\tan \delta = 0.003$, demonstrating their suitability for high-power ultrasonic transducer applications. The resulting ceramic exhibited excellent dielectric and piezoelectric properties, including $d_{33} = 455 \text{ pC/N}$, $Q_m = 1455$, and $\tan \delta = 0.003$, suitable for fabricating Langevin transducers for ultrasonic cleaning applications. Luo et al. (2022) added 0.3 wt.% Li_2CO_3 to the $\text{Pb}_{0.96}\text{Sr}_{0.04}(\text{Zr,Ti})_{0.7}(\text{Zn}_{1/3}\text{Nb}_{2/3})_{0.3}\text{O}_3$ (PZN–PZT) system and achieved good electrical characteristics with $d_{33} = 540 \text{ pC/N}$ and $k_p = 0.58$, while lowering the sintering temperature to 900°C . For commercial PZT (PZT-5H) doped with 0.2 wt.% Li_2CO_3 , the sintering temperature was successfully reduced to 980°C while maintaining a notably high d_{33} of 701 pC/N (Lin et al., 2023). Pu et al. (2022) doped 2.0 mol.% Li_2CO_3 into the PNN-PZT system and achieved an extremely high d_{33} (936 pC/N) and k_p (0.701) at a low sintering temperature of 960°C . The underlying mechanism involves Li^+ substitution at the B position of the perovskite network, promoting increased grain size and reducing the pinning effect of grain boundaries on domain wall mobility, thereby enhancing the piezoelectric effect (Pu et al., 2022). Nevertheless, most existing studies primarily focus on the role of Li_2CO_3 as a sintering aid, while its influence on defect chemistry, domain structure, and phase stability, particularly in complex multi-component systems combining hard and soft characteristics remains insufficiently understood. In addition, the interaction between Li_2CO_3 -induced liquid-phase sintering and defect-mediated domain engineering has not been systematically clarified.

Previous studies have demonstrated that the PSLZT–BMT hard piezoelectric ceramic system synthesized at 1150 °C exhibited a very wide range of composition tuning, from soft piezoelectric composition with $d_{33} = 640$ pC/N, $Q_m = 90$ to hard piezoelectric composition ($Q_m > 700$) when the BMT content reaches 0.024 mol (Van Thinh et al., 2023). Furthermore, the incorporation of PMS in the PSLZT–BMT matrix, along with Fe_2O_3 doping, provided an optimal balance between piezoelectric activity and mechanical strength, as shown by a very high mechanical quality factor ($Q_m = 2000$), while maintaining a $d_{33} = 420$ pC/N value and an extremely low dielectric loss ($\tan \delta = 0.003$) (Vuong et al., 2025). Despite these advantages, such systems still require relatively high sintering temperatures (~ 1150 °C), limiting their practical applications, especially in multilayer and co-fired devices. Therefore, the incorporation of Li_2CO_3 into the PSLZT–BMT–PMS system is expected to play a dual role: (i) promoting densification via liquid-phase-assisted sintering, thereby reducing the sintering temperature, and (ii) modifying defect chemistry and domain wall dynamics through Li-related defect formation. The synergistic interaction between liquid-phase sintering, defect dipoles, and phase instability near the morphotropic phase boundary (MPB) is anticipated to provide a pathway for simultaneously optimizing piezoelectric performance and mechanical robustness (Zhou et al., 2025).

Therefore, this study aims to systematically investigate the effects of Li_2CO_3 addition (0–1.0 wt.%) on the sintering behavior, phase structure, microstructure, and electromechanical properties of PSLZT–BMT–PMS ceramics. Particular emphasis is placed on elucidating the underlying mechanisms governing the interplay between densification, and defect chemistry, in order to establish an effective strategy for designing high-performance hard piezoelectric ceramics operating at reduced sintering temperatures.

2 Experimental

2.1 Material fabrication

The piezoelectric ceramic system with the nominal composition $[(\text{Pb}_{0.94}\text{Sr}_{0.05}\text{La}_{0.01})(\text{Zr}_{0.51}\text{Ti}_{0.49})\text{O}_3] - [\text{Bi}(\text{Mn}_{1/2}\text{Ti}_{1/2})\text{O}_3] - [\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3] + x$ wt.% Li_2CO_3 ($x = 0.0$ – 1.0), herein referred to as PSLZT–BMT–PMS + x wt.% Li_2CO_3 was synthesized using the conventional solid-state reaction technique. The raw materials consisted of high-purity starting oxides and carbonates, including PbO , ZrO_2 , TiO_2 , SrCO_3 , La_2O_3 , Bi_2O_3 , MnO_2 , Sb_2O_3 , and Li_2CO_3 (99.8% purity; Daejung, Korea). To compensate for PbO volatilization during high-temperature processing, an excess of 2 wt.% PbO was intentionally introduced. Initially, the PSLZT–BMT–PMS precursor mixture was homogenized via planetary ball milling in ethanol with zirconia media. The milling process was carried out for 24 h at a ball-to-powder mass ratio of 2:1. Subsequently, the slurry was dried and uniaxially pressed into disk-shaped pellets (diameter: 30 mm) under a pressure of 1500 kg/cm². The green compacts were calcined at 850 °C for 2 h with a heating rate of 5 °C/min, ensuring the completion of solid-state reactions. The calcined powders were then subjected to a second milling step under identical conditions to enhance homogeneity and reduce particle size. To ensure uniform additive distribution, the powders were mixed with varying amounts of Li_2CO_3 ($x = 0.0, 0.2, 0.4, 0.6, 0.8$, and 1.0 wt.%) for 30 min after drying. The resulting mixtures were pressed into pellets (diameter: 12 mm; thickness: 1.4 mm) using a uniaxial pressure of 500 kg/cm².

To minimize PbO volatilization during sintering, the samples were embedded in sacrificial powder of the same composition and placed in a semi-covered alumina crucible. Sintering was carried out in ambient air at temperatures ranging from 950 °C to 1150 °C for 2 h with a heating rate of 5 °C/min, followed by natural cooling to room temperature. Each composition was prepared in at least three independent batches to ensure reproducibility.

2.2 Characterization

The crystal structure of the ceramics was characterized by X-ray diffraction (XRD) using a D8-ADVANCE diffractometer. The microstructure and morphology were examined using a JEOL JSM-IT200 scanning electron microscope (SEM). The average grain size was determined using the linear intercept method based on SEM images, and grain size distributions were fitted using Gaussian functions. The bulk density was measured using the Archimedes method in an ethanol medium. The sintered samples were surface-polished, machined to standard piezoelectric dimensions, and coated with silver electrodes for electrical characterization. Following electrode deposition, the samples were poled in silicone oil under a DC electric field of 30 kV/cm at 120 °C for 30 minutes. These poling conditions were optimized to achieve stable polarization without dielectric breakdown. The poled samples were then rapidly cooled to room temperature and aged for 24 hours before measurement. The dielectric constant (ϵ) and dielectric loss ($\tan \delta$) were measured as a function of temperature using an LCR meter (HIOKI 3532) in the frequency range of 1–100 kHz. The electromechanical coupling coefficients (k_p and k_t) and mechanical quality factor (Q_m) were determined from impedance–phase frequency spectra according to IEEE piezoelectric standards. The piezoelectric coefficient d_{33} was directly measured using a ZJ-4B quasi-static d_{33} meter. The ferroelectric hysteresis (P – E) loops were recorded using the Sawyer–Tower circuit method. Measurement repeatability was confirmed by testing multiple samples from different batches, ensuring consistency of the reported results.

3 Results and discussion

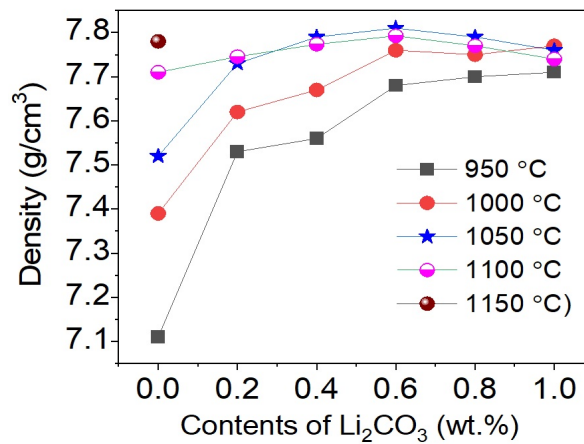


Figure 1: Bulk density of the ceramics as a function of Li_2CO_3 concentration, sintered at temperatures ranging from 950 °C to 1150 °C.

Fig. 1 illustrates the dependence of the bulk density (ρ) of the ceramic system $0.970[(\text{Pb}_{0.94}\text{Sr}_{0.05}\text{La}_{0.01})(\text{Zr}_{0.51}\text{Ti}_{0.49})\text{O}_3] - 0.024[\text{Bi}(\text{Mn}_{1/2}\text{Ti}_{1/2})\text{O}_3] - 0.006[\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3]$ on Li_2CO_3 content (0–1.0 wt.%) at sintering temperatures of 950–1150 °C. The results show that the ceramic density increases with increasing Li_2CO_3 content, reaching a maximum at 0.6 wt.%. However, when the content exceeds the optimal threshold, the density tends to saturate or slightly decrease at all investigated temperatures.

The undoped component exhibited a low ceramic density of 7.10 g/cm³ at the low sintering temperature of 950 °C, indicating an incomplete solid-phase reaction. With the addition of 0.2 wt.% Li_2CO_3 , the ceramic density increased to 7.52 g/cm³, signifying the role of Li_2CO_3 in enhancing sintering and promoting mass diffusion (Pu et al., 2022; Lin et al., 2023). Subsequently, the ceramic density increased with increasing Li_2CO_3 content, reaching 7.71 g/cm³ at a concentration of 1.0 wt.%. At 1000 °C, the ceramic density followed a similar increasing pattern

as at 950 °C, rising from 7.39 g/cm³ to 7.75 g/cm³ with increasing Li_2CO_3 content. Notably, the material achieved significantly higher densities at higher temperatures (1050–1100 °C), ranging from 7.75–7.80 g/cm³ at a Li_2CO_3 content of approximately 0.6 wt.%, demonstrating almost complete density conversion. This improvement is attributed to a liquid-phase-assisted sintering mechanism. During heating, Li_2CO_3 decomposes into Li_2O , which reacts with PbO to form a transient liquid phase, thereby enhancing particle rearrangement, grain boundary diffusion, and pore elimination. As a result, densification is significantly promoted even at reduced sintering temperatures. However, the ceramic density decreased slightly as the Li_2CO_3 content increased further. This is due to the evaporation of Li_2O at high temperatures, which generates oxygen vacancy defects and leads to the formation of micropores in the structure, thereby reducing the density. In addition, possible segregation of Li-rich species at grain boundaries may further hinder effective densification. It can be concluded from these results that adding an appropriate amount of Li_2CO_3 ($x = 0.6$ wt.%) and sintering at 1050 °C is the optimal condition to achieve the highest density of approximately 7.78 g/cm³, which is greater than that of the undoped sample sintered at 1150 °C. These results are consistent with recent reports on PZT systems and similar perovskite systems. In these studies, Li_2CO_3 is shown to be an effective sintering aid, reducing sintering temperature by 100–150 °C while still achieving high density and good electrical properties (Pu et al., 2022; Lin et al., 2023). Thus, integrating Li_2CO_3 addition with sintering temperature optimization is an effective strategy to enhance density and improve microstructure and ferro-piezoelectric properties of PSLZT–BMT–PMS materials.

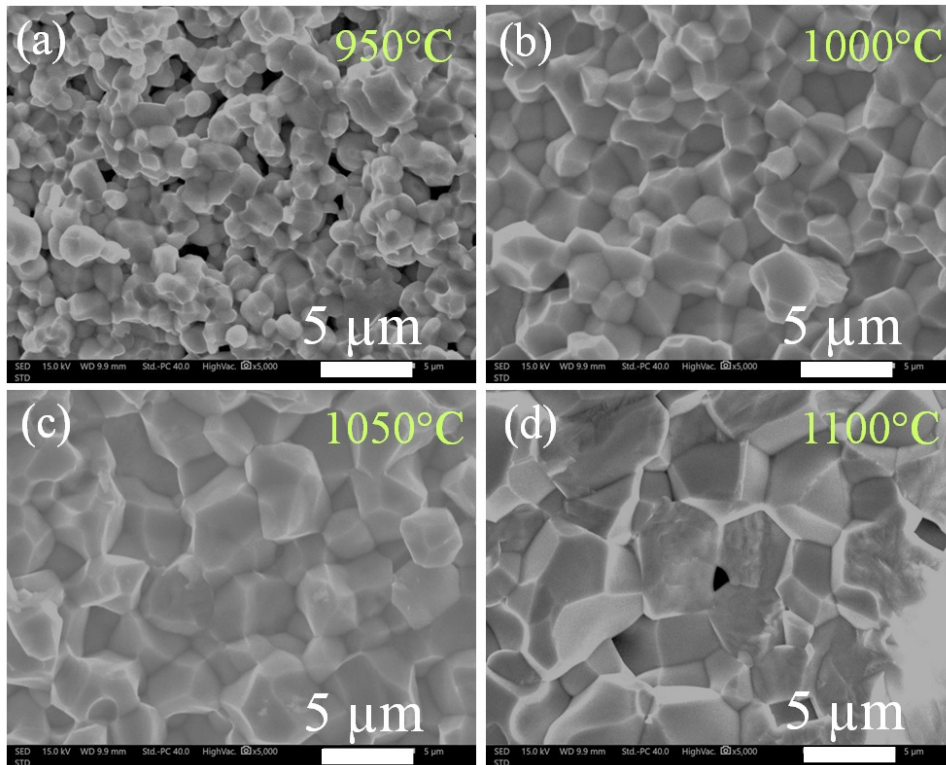


Figure 2: SEM micrographs of Li_2CO_3 -doped PSLZT–BMT–PMS ceramics sintered at various temperatures in the range of 950–1100 °C.

Fig. 2 shows the surface morphology of PSLZT–BMT–PMS ceramics sintered at temperatures in the range of 950–1100 °C, examined using SEM. The results indicate that the microstructure of the material changes significantly with increasing sintering temperature. At 950 °C, the ceramic sample exhibits numerous pores and small grains (Fig. 2(a)) and is not fully developed, indicating the onset of mass diffusion and grain rearrangement. As the temperature increases to 1000 °C (Fig. 2(b)), the pore density decreases significantly, grain boundaries become more

distinct, and grains begin to take on the polyhedral shape characteristic of perovskite structure. The grains become uniform and clearly defined at 1050 °C (Fig. 2(c)), indicating that the sintering process has reached an optimal density state. The uniform particle size and high density reflect the maximum ceramic density value in Fig. 1, indicating that the sintering process is most efficient in this temperature range. This result is similar to the study by Tiwari & Srivastava (2015), in which the addition of Li_2CO_3 formed a liquid phase that facilitated particle dissolution and migration, thereby improving the density process at low firing temperatures. Upon increasing the temperature to 1100 °C (Fig. 2(d)), the particles grew vigorously, resulting in an increased average size. Simultaneously, excessive particle growth and some secondary voids appeared between the particles. This usually occurs when the particle boundary diffusion rate exceeds the mass diffusion rate, resulting in an increased size difference between particles and a reduction in bulk density. This is similar to the report by Chung et al. (2004), in which high-temperature sintering adversely affects the microstructure and material properties.

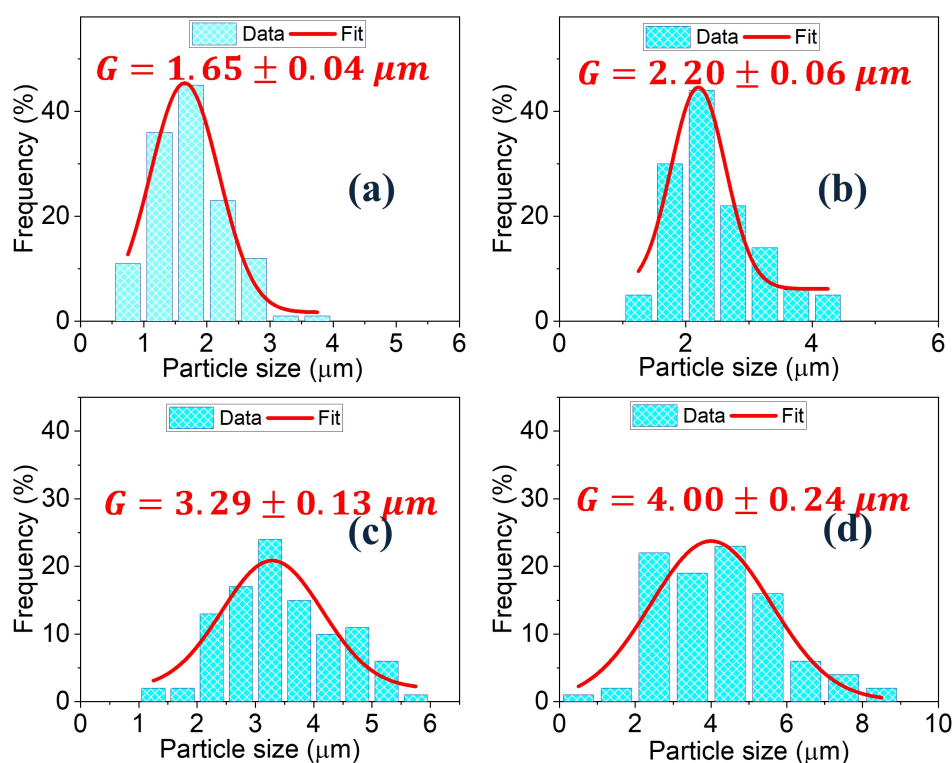


Figure 3: Grain size distribution of Li_2CO_3 -doped PSLZT-BMT-PMS ceramics sintered at various temperatures: (a) 950 °C, (b) 1000 °C, (c) 1050 °C, and (d) 1100 °C.

Fig. 3 presents the changes in grain size distribution of the PSLZT-BMT-PMS ceramic samples sintered at temperatures ranging from 950 to 1100 °C. As shown in Fig. 3, the average grain size G gradually increases with sintering temperature, from $1.65 \pm 0.04 \mu\text{m}$ at 950 °C (Fig. 3(a)) to $4.00 \pm 0.24 \mu\text{m}$ at 1100 °C (Fig. 3(d)). Simultaneously, the grain width also increases, indicating a broadening of the grain size spectrum with rising temperature. At 950 °C and 1000 °C (Fig. 3(a)-(b)), the grain size distribution is narrow in the range of 1.0–2.5 μm , indicating uneven grain growth in the early stages of sintering. The particle size distribution spectrum widens as the temperature reaches 1050 °C (Fig. 3(c)) and shifts towards larger sizes, with an average size of approximately $3.29 \pm 0.13 \mu\text{m}$. The particle growth becomes more pronounced as the temperature increases to 1100 °C (Fig. 3(d)), leading to particle size inhomogeneity and a significant expansion of the particle distribution. This behavior is attributed to anomalous particle growth, which occurs when the particle boundary migration rate exceeds the stability threshold due to the formation of a localized liquid phase from the decomposition of Li_2CO_3 .

into Li_2O (Siddiqui et al., 2017; Lin et al., 2023; Tiwari & Srivastava, 2015; Zhang et al., 2017a). This liquid phase formation reduces the particle boundary energy and promotes mass diffusion, leading to uncontrolled growth of some large particles.

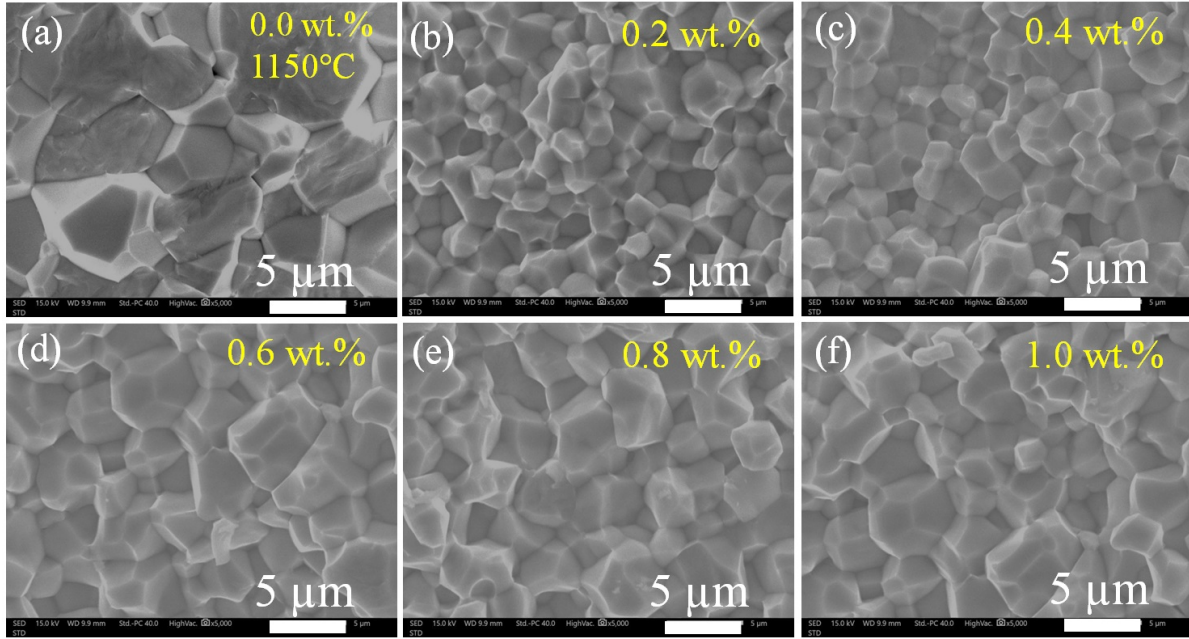


Figure 4: SEM images of PSLZT–BMT–PMS ceramics with different Li_2CO_3 contents: (a) 0.0 wt.% sintered at 1150 °C, (b) 0.2 wt.%, (c) 0.4 wt.%, (d) 0.6 wt.%, (e) 0.8 wt.%, and (f) 1.0 wt.% sintered at 1050 °C.

Fig. 4 displays the effect of Li_2CO_3 content on the microstructure of PSLZT-BMT-PMS ceramics. The undoped sample sintered at 1150 °C (Fig. 4(a)) exhibits large, irregular grains with evident inhomogeneity, indicating uncontrolled grain growth at high temperature. An experiment was conducted by adding Li_2CO_3 as the sintering aid at concentrations ranging from 0.2 wt. to 1.0 wt. and sintering at 1050 °C to compare with the undoped control sample sintered at 1150 °C. The results demonstrated that ceramics supplemented with a small amount of Li_2CO_3 from 0.2 to 0.6 wt.% and sintered at a lower temperature of 1050 °C exhibited a significantly finer, more uniform, and denser microstructure (Fig. 4(b)–(d)). At 0.6 wt.%, the particles have a defined shape and sharp grain boundaries, indicating a shift to an optimally controlled grain growth mechanism. This improvement demonstrates that Li_2CO_3 enables microstructural refinement at reduced sintering temperature, which cannot be achieved in the undoped system even at 1150 °C.

The enhanced microstructural control is attributed to liquid-phase-assisted sintering induced by Li_2CO_3 decomposition, which lowers grain boundary energy and promotes uniform mass transport. As a result, grain growth proceeds in a controlled manner without excessive coarsening (Siddiqui et al., 2017; Pu et al., 2022; Zhang et al., 2022; Fan et al., 2014; Zhao & Bian, 2023). However, when the Li_2CO_3 content increases to 0.8–1.0 wt. (Fig. 4(e)–(f)), localized grain coarsening becomes evident, with large grains embedded within a finer matrix. This suggests the onset of abnormal grain growth, likely caused by excessive liquid phase and increased grain boundary mobility, leading to preferential growth of certain grains (Quang & Vuong, 2022; Ma et al., 2024; Tu et al., 2025). However, this growth rate remained significantly lower than that of the undoped sample at 1150 °C, suggesting the effectiveness of Li_2CO_3 in maintaining a stable microstructure at lower sintering temperatures.

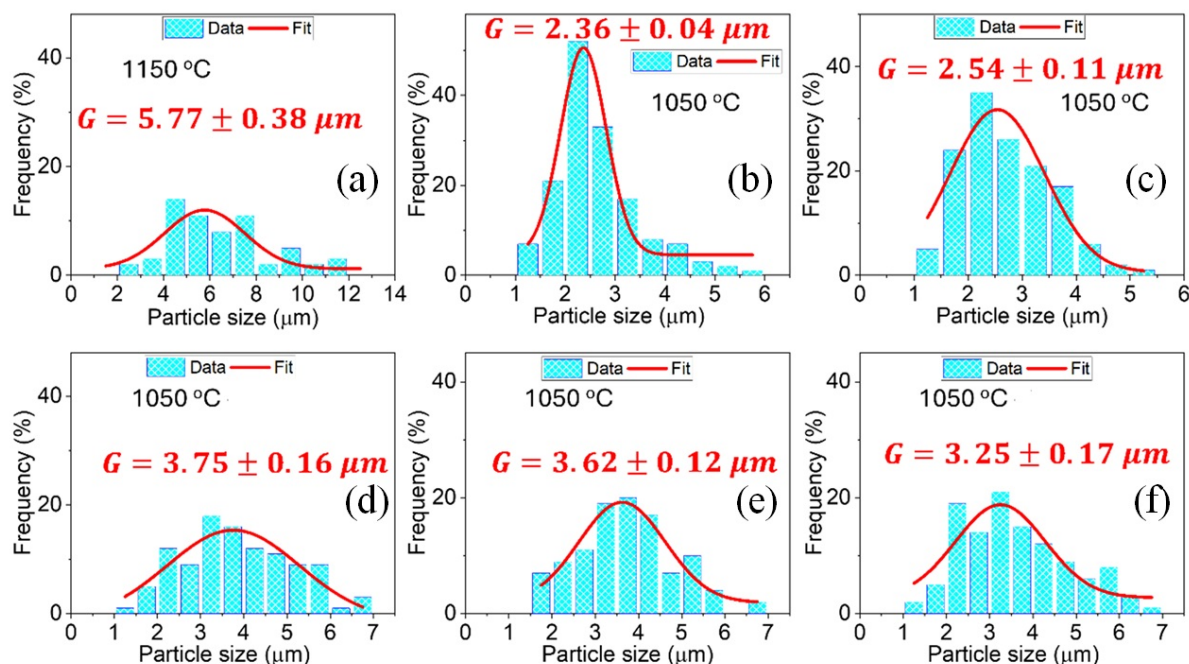
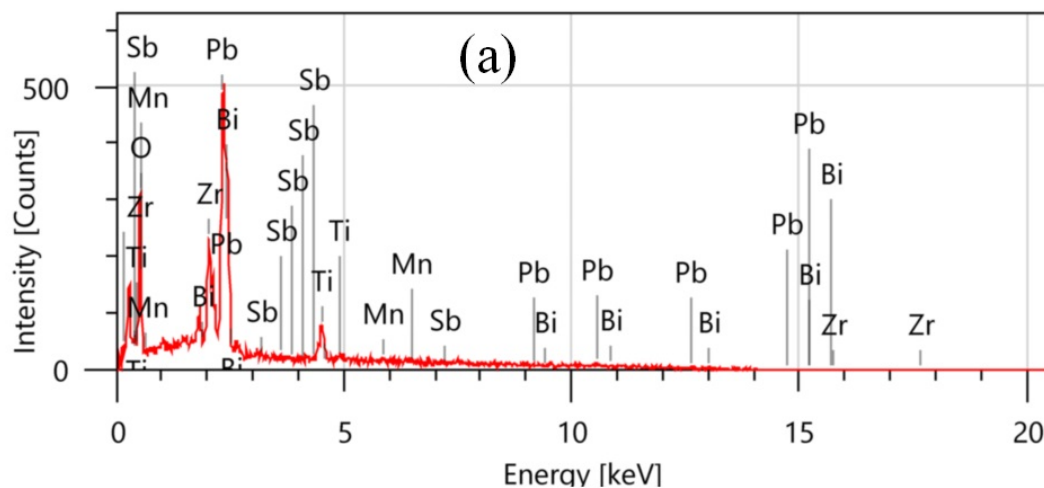


Figure 5: Grain size distributions of PSLZT-BMT-PMS ceramics with different Li_2CO_3 contents: (a) 0.0 wt.% sintered at 1150 °C, (b) 0.2 wt.%, (c) 0.4 wt.%, (d) 0.6 wt.%, (e) 0.8 wt.%, and (f) 1.0 wt.% sintered at 1050 °C.



Element	Line	Mass%	Atom%
O	K	19.05±0.44	68.05±1.57
Ti	K	5.44±0.29	6.49±0.34
Mn	K	0.41±0.17	0.43±0.18
Zr	L	12.25±0.45	7.68±0.28
Sb	L	0.10±0.25	0.05±0.12
Pb	M	59.61±1.08	16.45±0.30
Bi	M	3.14±0.74	0.86±0.20
Total		100.00	100.00

Figure 6: (a) Energy-dispersive X-ray spectroscopy (EDS) spectrum of the Li_2CO_3 -doped PSLZT-BMT-PMS ceramics, and (b) the elemental composition obtained from energy-dispersive X-ray spectroscopy (EDS) analysis.

Fig. 5 presents the results of the Gaussian particle size distribution analysis and the determination of the mean particle size G as a function of Li_2CO_3 content from the SEM images. The average grain size increases from $2.36 \mu\text{m}$ to $\sim 3.75 \mu\text{m}$ as the Li_2CO_3 content increases from 0.2 to 0.6 wt.%, accompanied by a relatively narrow and symmetric distribution, indicating uniform grain growth (Fig. 5(b)-(d)). At higher concentrations (>0.6 wt.%), the grain size distribution broadens and becomes less uniform, confirming the transition to abnormal grain growth behavior. (Fig. 5 (e), (f)) (Vuong & Gio, 2013). Thus, these results indicate that an optimal Li_2CO_3 content (~ 0.6 wt.%) enables controlled grain growth, resulting in a dense and homogeneous microstructure. In contrast, excessive Li_2CO_3 leads to microstructural instability due to enhanced grain boundary kinetics and liquid-phase effects (Zhang et al., 2022; Vuong & Gio, 2013).

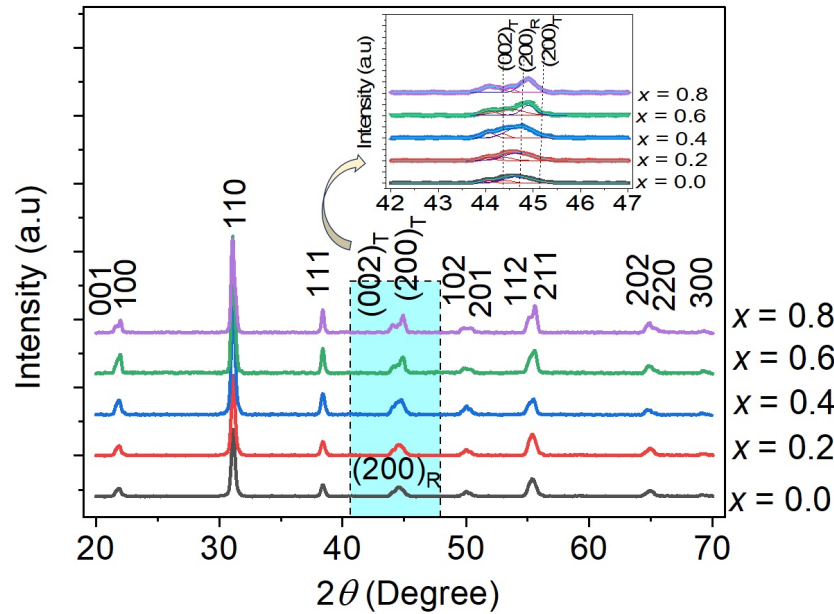


Figure 7: X-ray diffraction (XRD) patterns of PSLZT-BMT-PMS ceramics with various Li_2CO_3 doping concentrations.

Fig. 6(a) presents the EDS spectrum of the Li_2CO_3 -modified PSLZT-BMT-PMS ceramic, confirming the presence of the main constituent elements Pb, Zr, Ti, and O, along with dopant elements Sr, La, Mn, Sb, and Bi. The detection of both A-site (Sr^{2+} , La^{3+} , Bi^{3+}) and B-site (Zr^{4+} , Ti^{4+} , $\text{Mn}^{2+}/\text{Mn}^{3+}$, Sb^{5+}) elements indicates the successful incorporation of complex perovskite modifiers into the host lattice. Quantitative results (Fig. 6(b)) show that the mass percentages of the main elements are Pb (56.26%), Zr (13.39%), Ti (6.66%), and O (17.09%). Their corresponding atomic percentages are 16.09%, 8.7%, 8.24%, and 63.30%, which are close to the nominal percentages in the PZT composition. Doping elements, including Bi (1.76 wt.%), Mn (0.78 wt.%), Sb (0.73 wt.%), Sr (2.17 wt.%), and La (1.16 wt.%), were detected in low amounts. The presence of Bi, Mn, and Sb is associated with the BMT and PMS components, which play a critical role in defect engineering. In particular, acceptor-type dopants such as Mn and Sb are known to induce oxygen vacancies, forming defect dipoles that interact with ferroelectric domains. This interaction effectively pins domain walls, leading to increased mechanical quality factor (Q_m) and characteristic "hard" piezoelectric behavior (Liu et al., 2024). However, the element Li was not observed in the EDS spectrum due to its very small mass percentage and its role as a sintering aid (Quang & Vuong, 2022).

Fig. 7 presents the X-ray diffraction patterns of PSLZT-BMT-PMS ceramics with varying Li_2CO_3 content ($x = 0.0$ – 0.8 wt.%). The results show that all the samples exhibit characteristic diffraction peaks corresponding to the perovskite structure, specifically for the (100), (110),

(111), (200), (210), (211), (220), and (300) planes. As the doping content increases ($x = 0.2$ – 0.8 wt.%), significant shifts and intensity variations were observed in the diffraction peaks, particularly in the $(200)_R$ region, reflecting a transformation of the crystal structure (Zhang et al., 2022; Vuong & Gio, 2013). The insert of Fig. 7 shows the X-ray diffraction spectra of the magnified ceramic samples in the 2θ region of 42° – 47° . For the undoped sample $x = 0.0$ wt.%, the sample exhibits a dominant rhombohedral phase structure characterized by the $(200)_R$. Notably, the $(200)_R$ peak splits into two peaks, $(200)_R$ and $(200)_T$, as the Li_2CO_3 content increases to $x = 0.4$ – 0.6 wt.%. At $2\theta = 45^\circ$, three distinct peaks $(002)_T/(200)_R/(200)_T$ are observed, indicating the coexistence of two tetragonal–rhombohedral (T – R) phases at the morphological phase boundary (MPB). The $(200)_*T$ peak becomes dominant and shifts towards larger angles at $x > 0.6$ wt.%, showing a gradual shift towards the dominant tetragonal phase (Pu et al., 2022). This is attributed to the substitution of Li^+ ions with a larger radius $R_{\text{Li}^+} = 0.740 \text{ \AA}$ into the B-site, replacing ions with smaller radii, such as $R_{\text{Zr}^{4+}} = 0.720 \text{ \AA}$ and $R_{\text{Ti}^{4+}} = 0.605 \text{ \AA}$ (Hou et al., 2007). The morphological phase boundary region (MPB) was observed in the Li_2CO_3 compositional range of $x = 0.4$ – 0.6 wt.%. This range is important for piezoelectric materials, as the coexistence of rhombohedral-tetragonal crystal phases increases the mobility of the polarization vector component, thereby significantly enhancing the dielectric and piezoelectric properties of PSLZT-BMT-PMS ceramics (Pu et al., 2022).

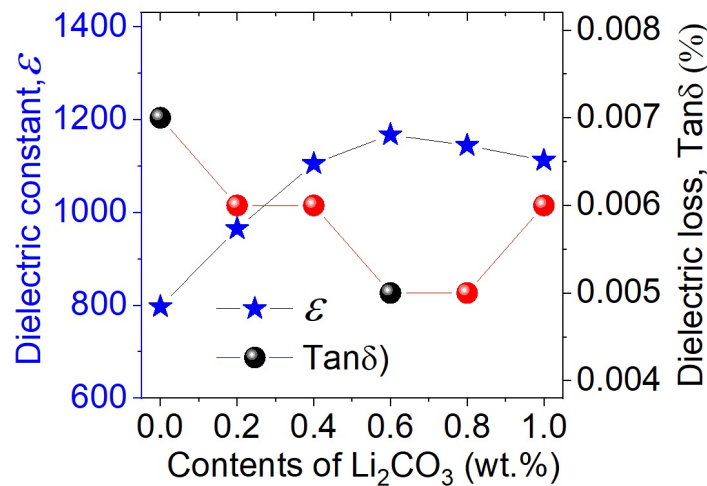


Figure 8: Dielectric constant and dielectric loss as a function of Li_2CO_3 concentration for PSLZT–BMT–PMS ceramics.

Fig. 8 shows the dependence of dielectric constant (ϵ) and dielectric loss ($\tan \delta$) on Li_2CO_3 content (wt.%) in the PSLZT-BMT-PMS ceramic sample after sintering, measured at 1 kHz. The results show that for the undoped sample ($x = 0.0$ wt.%), the dielectric constant ϵ is approximately 800. Notably, ϵ gradually increases with doping content, reaching a maximum of nearly 1196 at $x = 0.6$ wt.%, and decreases slightly when x exceeds 0.6 wt.%. The enhancement in dielectric properties at intermediate compositions can be attributed to the combined effects of improved densification, reduced porosity, and optimized grain structure, which collectively enhance polarization response. More importantly, this behavior correlates strongly with the formation of a morphotropic phase boundary (MPB), as identified in Fig. 7. The coexistence of rhombohedral and tetragonal phases facilitates polarization rotation under an applied electric field, thereby significantly increasing dielectric permittivity (Hou et al., 2007). The dielectric loss ($\tan \delta$) of the sample without sintering aid is approximately 0.007. It decreases to the smallest value of 0.005 at $x = 0.6$ wt.%, and then increases again as x increases to 1.0 wt.%. The reduction in dielectric loss at the optimal composition indicates a decrease in defect-related energy dissipation, suggesting improved microstructural homogeneity and reduced defect concentration.

However, at higher Li_2CO_3 contents (> 0.6 wt.%), the increase in $\tan \delta$ may be associated with defect accumulation and microstructural inhomogeneity, such as grain boundary segregation and residual porosity (Siddiqui et al., 2017).

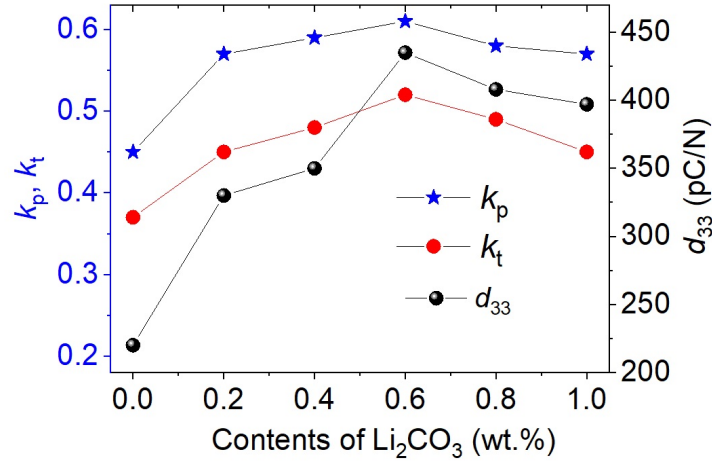


Figure 9: Radial electromechanical coupling coefficient (k_p), thickness electromechanical coupling coefficient (k_t), and piezoelectric coefficient (d_{33}) as functions of Li_2CO_3 concentration.

Fig. 9 illustrates the dependence of electromechanical parameters, including the radial electromechanical coupling coefficient (k_p), the longitudinal coupling coefficient (k_t), and the piezoelectric constant (d_{33}), on the Li_2CO_3 content (wt.%) in PSLZT-BMT-PMS ceramics. The results show that all three parameters increased significantly with Li_2CO_3 addition up to approximately 0.6 wt.%, and then decreased slightly as the doping concentration exceeded 0.6 wt.%. Specifically, the radial electromechanical coupling coefficient (k_p) increased from 0.45 at $x = 0.0$ wt.% to a maximum value of 0.60 at $x = 0.6$ wt.%, reflecting a significantly improved radial electromechanical energy conversion. Similarly, the thickness-oriented electromechanical coupling coefficient (k_t) also reached a maximum value of 0.53 at $x = 0.6$ wt.%. The piezoelectric constant (d_{33}) increased from 220 pC/N at $x = 0.0$ wt.% to approximately 430 pC/N at $x = 0.6$ wt.% and then decreased with a continued increase in doping concentration. This enhancement can be understood in terms of both intrinsic and extrinsic contributions. The MPB structure enhances intrinsic polarization rotation, while the optimized microstructure (high density and uniform grain size) facilitates domain wall motion, contributing to extrinsic piezoelectric response. The combined effect results in significantly improved electromechanical performance at the optimal composition (Siddiqui et al., 2017; Zhang et al., 2022). However, when the Li_2CO_3 content exceeds 0.6 wt.%, the doping exceeds the maximum solubility threshold, causing the electromechanical and piezoelectric coupling parameters to degrade. This degradation may result from the space charges located at the grain boundaries caused by the excess Li accumulation, and space charges also have the effect of pinning domain walls, causing a reduction in piezoelectric properties similar to that reported by (Zhang et al., 2022). In addition, deviation from the MPB composition toward a tetragonal-dominant phase further limits polarization rotation, contributing to the observed decrease in properties. The results demonstrate that the optimal Li_2CO_3 content (~ 0.6 wt.%) provides a synergistic balance between phase structure (MPB), microstructure (density and grain size), and defect chemistry, leading to enhanced dielectric and piezoelectric performance.

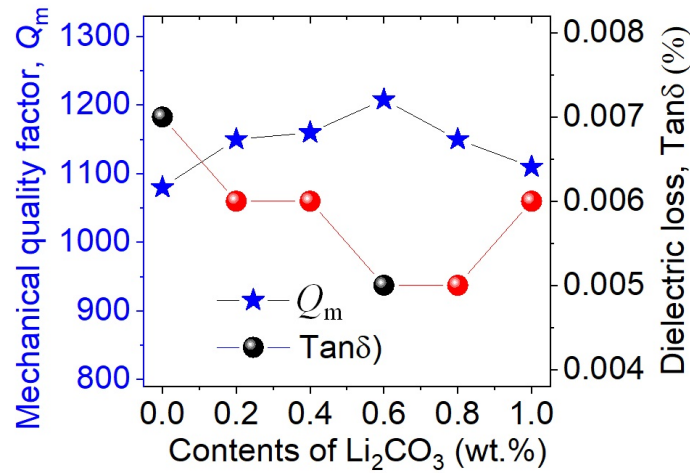


Figure 10: Mechanical quality factor (Q_m) and dielectric loss ($\tan \delta$) as functions of Li_2CO_3 concentration for PSLZT-BMT-PMS ceramics measured at room temperature.

Fig. 10 shows the variation of the mechanical quality factor (Q_m) and dielectric loss ($\tan \delta$) with Li_2CO_3 content ($x = 0\text{--}1.0$ wt.%). The results show that the Q_m value gradually increases from approximately 1070 to a maximum value of approximately 1220 at $x = 0.6$ wt.% as the Li_2CO_3 content increases from 0 to 0.6 wt.%. Q_m decreases slightly as Li_2CO_3 continues to increase to 1.0 wt.%. Conversely, the dielectric loss ($\tan \delta$) of the PSLZT-BMT-PMS material before Li_2CO_3 addition is approximately 0.007, and decreases to a minimum value of 0.005 as the Li_2CO_3 content increases to $x = 0.6$ wt.%. The simultaneous increase in Q_m and decrease in $\tan \delta$ at intermediate compositions indicates enhanced “hard” piezoelectric behavior. This can be attributed to optimized defect chemistry and microstructural homogeneity. In particular, the presence of acceptor-type dopants (Mn, Sb) is known to generate oxygen vacancies, forming defect dipoles that interact with ferroelectric domains. At the optimal Li_2CO_3 content, these defect dipoles are effectively stabilized within a dense and uniform microstructure, leading to strong domain wall pinning and reduced energy dissipation, thereby increasing Q_m and lowering dielectric loss. However, at higher Li_2CO_3 contents (> 0.6 wt.%), the slight degradation of Q_m and increase in $\tan \delta$ can be associated with excess defect formation and grain boundary effects. The accumulation of Li-related species or space charges at grain boundaries may disrupt defect dipole alignment and introduce additional loss mechanisms, resulting in reduced mechanical quality and increased dielectric loss (Siddiqui et al., 2017; Vuong & Gio, 2013).

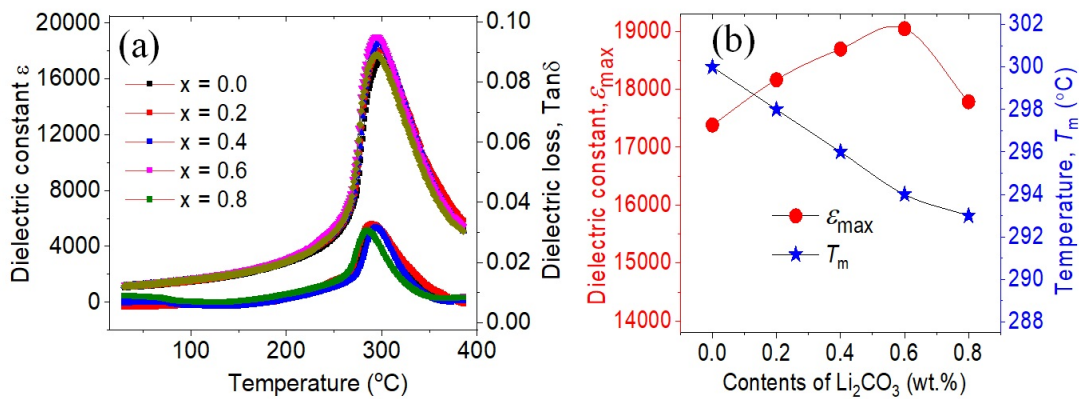


Figure 11: (a) Temperature-dependent dielectric constant (ϵ_r) and dielectric loss ($\tan \delta$) of the ceramics, and (b) the maximum dielectric constant ($\epsilon_{\max}$) and phase transition temperature (T_m) as functions of Li_2CO_3 concentration.

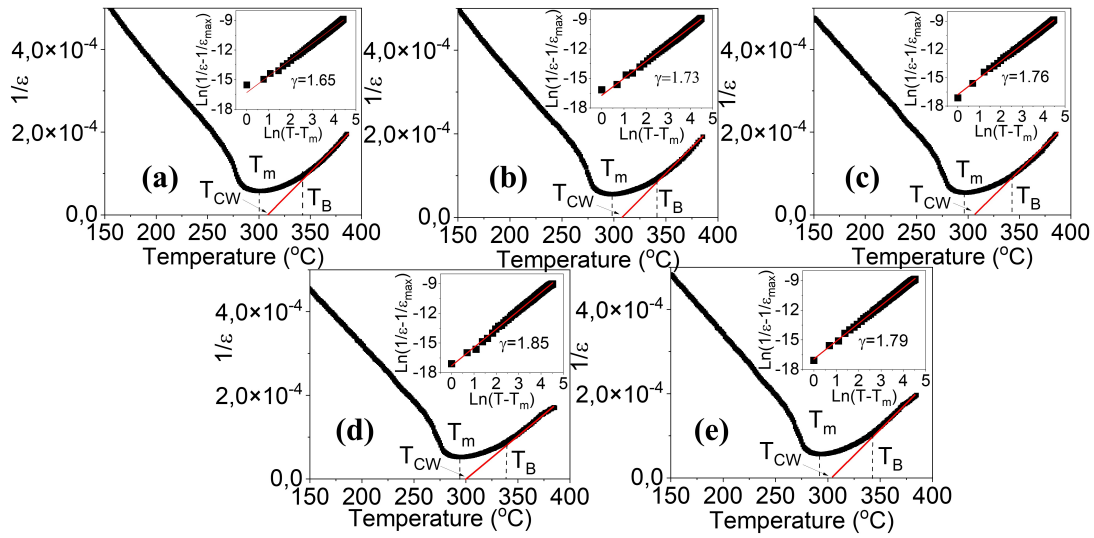


Figure 12: Curie–Weiss plots of PSLZT–BMT–PMS ceramics with different Li_2CO_3 concentrations: (a) $x = 0.0$ wt.%, (b) $x = 0.2$ wt.%, (c) $x = 0.4$ wt.%, (d) $x = 0.6$ wt.%, and (e) $x = 0.8$ wt.%.

Fig. 11(a) shows the dependence of dielectric constant (ϵ) and dielectric loss ($\tan \delta$) on temperature for PZT ceramic samples doped with different amounts of Li_2CO_3 khác nhau, measured at 1 kHz. The results indicate that all samples have a peak in ϵ at the ferroelectric-paraelectric phase transition temperature (T_m) over a wide temperature range, characteristic of relaxor materials (Pu et al., 2022). Simultaneously, the corresponding dielectric constant peak value $\tan \delta_{\max}$ remains low (< 0.035), indicating small dielectric loss consistent with the characteristics of hard piezoelectric ceramics. The relaxor behavior can be attributed to compositional disorder at both A- and B-sites, leading to the formation of polar nano-regions (PNRs) and diffuse phase transitions. Figure 11(b) shows the variation of $\epsilon_{\max}$ and T_m with Li_2CO_3 content in PSLZT–BMT–PMS ceramics. The dielectric constant $\epsilon_{\max}$ increases with increasing Li_2CO_3 content, reaching a maximum of approximately 19050 at 0.6 wt.%, and then decreases with increasing flux. The enhancement of $\epsilon_{\max}$ is associated with improved domain response and increased grain size, while the subsequent decrease is related to structural and defect inhomogeneity at higher Li_2CO_3 concentrations (Vuong & Gio, 2013). In contrast, T_m tends to decrease linearly from 300 °C to 293 °C as Li_2CO_3 content increases to 0.8 wt.%. This reduction in T_m can be associated with lattice destabilization induced by Li incorporation into the perovskite structure. According to Hou et al. (Hou et al., 2007), the radius of the Li^+ ion ($R_{\text{Li}^+} = 0.74 \text{ \AA}$) is similar to that of Zr^{4+} and Ti^{4+} ($R_{\text{Zr}^{4+}} = 0.720 \text{ \AA}$, $R_{\text{Ti}^{4+}} = 0.605 \text{ \AA}$), which can substitute into the B-sites of the BO_6 octahedron. The Li^+ ions entering the perovskite crystal lattice affect the stability of the BO_6 octahedron because the bond energy of Li–O ($\approx 333.5 \text{ kJ/mol}$) is significantly lower than that of other B–O bonds (Zr–O: 776.1 kJ/mol, Ti–O: 672.4 kJ/mol), leading to a reduction in the phase transition temperature T_m . Additionally, increased compositional disorder further contributes to the diffuse phase transition behavior.

The inverse dielectric constant of the Li_2CO_3 -doped PSLZT–BMT–PMS ceramics, with concentrations ranging from 0.0 wt.% to 0.8 wt.%, as a function of temperature is presented in Fig. 12. The dielectric behavior above the phase transition temperature ($T > T_m$) was analyzed using the modified Curie–Weiss law (Vuong & Gio, 2013):

where $\epsilon_{\max}$ is the maximum value of the dielectric constant at the phase transition temperature T_m , γ is the degree of diffuseness, and C is the Curie constant. The upper-right insets of Fig. 12(a–f) display the values of the diffuseness coefficient (γ), which were evaluated by plotting $\ln(1/\epsilon - 1/\epsilon_{\max})$ as a function of $\ln(T - T_m)$ at a frequency of 1 kHz. The extracted γ values increase from 1.65 ($x = 0.0$ wt.%) to a maximum of 1.85 at $x = 0.6$ wt.%, followed by a slight

decrease to 1.79 at $x = 0.8$ wt.% (Table 1). This non-monotonic evolution indicates that Li_2CO_3 incorporation initially enhances the relaxor character of the system, before inducing a partial suppression at higher concentrations.

From a mechanistic perspective, the increase in γ is attributed to the enhanced structural and chemical disorder induced by multi-site substitution. Specifically, the coexistence of A-site substitution (La^{3+} , Sr^{2+}), B-site complex ions ($\text{Mn}^{3+}/\text{Sb}^{5+}$), and the incorporation of Li^+ ions with a relatively small ionic radius and weak bonding strength introduces local compositional fluctuations and random electric fields. These effects promote the formation of polar nanoregions (PNRs), which are responsible for the diffuse phase transition behavior (Hou et al., 2007). At higher Li_2CO_3 contents (> 0.6 wt.%), the reduction in γ suggests a deviation from the optimal disorder state. This behavior can be rationalized in terms of defect chemistry and solubility limits. Once the solubility threshold of Li^+ is exceeded, excess Li tends to segregate at grain boundaries rather than being incorporated into the lattice. Such segregation leads to local compositional inhomogeneity and reduced lattice randomness within grains, thereby weakening the relaxor character. Additionally, the accumulation of Li-related defects at grain boundaries can restrict domain-wall dynamics and reduce spatial polarization fluctuations, further contributing to the decrease in γ .

Table 1: Dielectric parameters of Li_2CO_3 -doped PSLZT-BMT-PMS ceramics.

Sample (wt.%)	T_m (°C)	T_B (°C)	T_{C-W} (°C)	$\varepsilon_{\max}$	γ	$C (\times 10^7)$
0.0	300	345	309	17374	1.65	1.24
0.2	298	343	307	18061	1.73	1.80
0.4	296	340	304	18588	1.76	1.93
0.6	294	338	300	19050	1.85	3.07
0.8	293	343	304	17706	1.79	2.08

The results of the analysis of the parameters T_m , T_B , T_{C-W} , and the Curie constant (C) obtained from Fig. 12 for different Li_2CO_3 concentrations are presented in Tab. 1. It can be observed that the values of T_m exhibit a slight downward shift with increasing Li_2CO_3 content, which can be attributed to the reduced lattice stability associated with Li-O bonding. In contrast, the Burns temperature (T_B) and Curie-Weiss temperature T_{C-W} show an opposite trend relative to γ , indicating that the degree of polar nanoregion formation and their thermal stability are strongly influenced by defect-induced lattice disorder. The Curie constant (C), ranging from 1.24 to 3.07, reflects the overall polarizability of the system and shows a composition-dependent variation associated with both grain size evolution and lattice distortion. The relatively high C values further confirm the strong dielectric response near the phase transition region.

4 Conclusions

In conclusion, this study reports the successful synthesis of Li_2CO_3 -doped PSLZT-BMT-PMS piezoelectric ceramic system using conventional solid-state methods. The incorporation of Li_2CO_3 effectively reduced the sintering temperature to 1050 °C through a liquid-phase-assisted densification mechanism, while simultaneously influencing the microstructure and phase evolution. In conclusion, Li_2CO_3 -modified PSLZT-BMT-PMS piezoelectric ceramics were successfully synthesized via a conventional solid-state route. The incorporation of Li_2CO_3 effectively reduced the sintering temperature to 1050 °C through a liquid-phase-assisted densification mechanism, while simultaneously influencing the microstructure and phase evolution. An optimal composition of 0.6 wt.% Li_2CO_3 exhibited a balanced combination of properties, including a density of 7.78 g/cm³, dielectric constant (ε) of 1196, low dielectric loss ($\tan \delta$) of 0.005, electromechanical coupling coefficients ($k_p = 0.60$, $k_t = 0.53$), piezoelectric coefficient ($d_{33} = 430$ pC/N), and mechanical quality factor ($Q_m = 1220$). These enhancements are attributed to

the synergistic effects of improved densification, controlled grain growth, and the formation of a rhombohedral–tetragonal phase coexistence near the morphotropic phase boundary (MPB). Compared with conventional Li_2CO_3 -modified PZT systems, the present composition provides a favorable trade-off between piezoelectric response and mechanical quality factor, which is particularly relevant for high-power applications. However, the improvement is limited by the solubility threshold of Li^+ (~ 0.6 wt.%), beyond which defect accumulation and grain boundary segregation lead to property degradation.

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Data availability

The raw data supporting the conclusions of this article will be made available by the authors on reasonable request.

Conflict of interests

The authors declare that they have no conflict of interest.

Contributions

Experimental design, investigation, and writing the original manuscript by **Vo Thanh Tung**. Conceptualization, building the experimental method, data analysis, and supervision by **Nguyen Van Thinh**. Data analysis, investigation, and editing by **Le Dai Vuong**. Theoretical analysis, review, and editing by **Do Viet On**. All authors read and approved the final manuscript.

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