

Review

Perovskite-Layered Ultra-High-Temperature Piezoelectric Ceramics: Research Progress in Structural Origins, Compositional Design, and Fabrication Processes

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ABSTRACT: Perovskite-layer structured (PLS) piezoelectric ceramics, featuring ultrahigh Curie temperatures (T_C), high electrical resistivity (ρ), and excellent thermal stability, are promising for sensing above 600 °C. However, poor densifiability, difficult poling, and low piezoelectric coefficients (d_{33}) still limit their practical applications. Focusing mainly on advances since 2018, this review critically evaluates recent progress in the structural understanding, compositional design, and processing of PLS ceramics. Improved control of lattice distortion, polarization switching, densification, and grain orientation has increased d_{33} to approximately 3–4 pC/N in $\text{Sr}_2\text{Nb}_2\text{O}_7$ -based ceramics and approximately 9 pC/N in $\text{La}_2\text{Ti}_2\text{O}_7$ -based systems. Nevertheless, *in situ* high-temperature piezoelectric data, long-term stability studies, and device-level validation remain limited. Further progress requires synergistic composition-processing optimization and standardized high-temperature evaluation to advance PLS ceramics toward practical ultrahigh-temperature sensing applications.

Keywords: Perovskite-layer structured; Ultrahigh-temperature piezoelectric ceramics; Piezoelectric coefficient; Densification

1. Introduction

Piezoelectric ceramics, owing to their electromechanical conversion capability, are widely used in sensors, actuators, and transducers [1–5]. With increasing demand for vibration and condition monitoring in aerospace engines, gas turbines, nuclear reactors, automotive systems, and other high-temperature industrial equipment, piezoelectric materials capable of stable operation at 600 °C and above are increasingly required. Such applications demand not only a high piezoelectric response but also a high T_C , high electrical resistivity, and excellent thermal stability [6,7].



In piezoelectric materials, a trade-off generally exists between T_C and d_{33} . Currently, widely used lead zirconate titanate (Pb(Zr, Ti)O₃, PZT)-based piezoelectric ceramics exhibit excellent piezoelectric properties, with d_{33} values of 300–700 pC/N, but their T_C is limited to 300–400 °C, making them primarily suitable for sensors operating below 200 °C. Although bismuth-layered structure piezoelectric ceramics possess relatively high T_C values of 500–940 °C, their d_{33} values are generally low, typically around 20 pC/N, and they are mainly applied to sensors with operating temperatures of approximately 400–500 °C [8]. Other high-temperature piezoelectric materials, such as AlN, LiNbO₃, and langasite, also exhibit favorable thermal stability. Langasite is non-ferroelectric and remains phase-stable to very high temperatures without thermal depoling, but the high cost of single-crystal growth limits its widespread application. AlN and LiNbO₃ are likewise limited by relatively low piezoelectric coefficients or reduced electrical resistivity at elevated temperatures. Table 1 compares the representative piezoelectric and high-temperature properties of these material systems. Therefore, existing high-temperature piezoelectric ceramics remain unable to meet the requirements for long-term stable operation in ultrahigh-temperature vibration sensors at 600 °C and above, which has, to some extent, hindered the development of such sensors.

PLS ultrahigh-temperature piezoelectric ceramics, with ultrahigh T_C (>1300 °C), high electrical resistivity, and excellent thermal stability, are promising candidates for sensing above 600 °C [9–11]. However, their restricted polarization direction, high coercive field ($E_C > 100$ kV/cm), and strong crystallographic anisotropy lead to incomplete poling, low piezoelectric coefficients ($d_{33} < 10$ pC/N), and poor densification during conventional sintering [12,13]. These limitations hinder their practical application in high-temperature sensing. Recent advances in textured piezoelectric ceramics show that combining compositional design with texture engineering can effectively enhance piezoelectric performance, providing useful guidance for PLS ceramics [14].

In our previous review published in 2018 [1], we identified poor sinterability, low piezoelectric response, and unclear ferroelectric phase-transition mechanisms as major challenges in PLS ceramics. The 2024 review [15] shares several foundational topics with the present work, including crystal structure, synthesis, fabrication, microstructure, electrical properties, and high-temperature applications. However, key questions remained open regarding powder-densification-property relationships, the control and actual benefit of self-texturing, the effects of composition and defects on polarization and domain behavior, and direct high-temperature piezoelectric response. Building on these studies, the present review incorporates evidence up to 2026 and highlights several advances: BO₆ octahedral rotations/tilting coupled with cation displacements as the structural origin of spontaneous polarization; the dual role of defects in intrinsic polarization and domain-wall mobility; a shift toward mechanism-guided multi-element doping and isostructural solid-solution design; and the coupled effects of densification, texture, poling, and high-temperature resistivity on d_{33} .

To make this intellectual progression more explicit, Table 2 compares the understanding established by 2018 with key post-2018 advances and remaining issues across six research aspects. This comparison highlights not only improvements in reported properties but also the shift from empirical modification toward a more mechanism-guided understanding of composition-structure-processing-property relationships in PLS ceramics.

Table 1. Comparison of representative high-temperature piezoelectric material systems [compiled by the authors from the cited primary literature].

Material System	d_{33} (pC/N)	T_C (°C)	ρ at 600 °C ($\Omega\cdot\text{cm}$)	T_p (°C)	Ref.
PZT ceramics	300–700	300–400	N/A	200–300	[2,6]
Aurivillius ceramics	~20	500–940	10^5 – 10^7	500–800	[16–18]
LiNbO ₃	~6	1150–1210	10^5 – 10^6	500–700	[19,20]
AlN	4.5–6.4	N/A	N/A	N/A	[21]
Langasite	N/A	N/A	10^5 – 10^6	N/A	[2]
PLS ceramics	1–9	1300–1450	$\sim 10^7$	1000–1200	[1,22–24]

N/A: not applicable or directly comparable. AlN and langasite are non-ferroelectric and therefore have no T_C or T_p , whereas 600 °C exceeds the T_C range of PZT; T_p : thermal depoling temperature.

Table 2. Evolution of understanding in PLS piezoelectric ceramics since the 2018 review [compiled by the authors from the cited literature as an original synthesis].

Research Aspect Understanding by 2018		Post-2018: Confirmation/Extension/Qualification
Origin of Spontaneous Polarization	Ultrahigh T_C and ferroelectricity were established, but the microscopic origin of polarization remained incompletely understood.	Confirmation: Structural mechanisms of ferroelectricity are further supported. Extension: Specific octahedral rotation/tilting modes, coupled cation displacements, and strain effects on polarization were further elucidated [25–27]. Qualification: Quantitative links to domain behavior and d_{33} remain unclear.
	A-/B-site substitution provided only limited d_{33} .	Confirmation: A-/B-site substitution can enhance piezoelectric response. Extension: Multi-element doping regulates local structural heterogeneity, polarization switching, and domain-wall contributions [22,28,29]. Qualification: Piezoelectric enhancement must still be balanced against phase stability, leakage, and T_C .
Solid-solution design	Secondary components were mainly introduced to improve densification and electrical properties.	Confirmation: Solid-solution formation improves electrical properties. Extension: Isostructural PLS solid solutions yield substantially enhanced d_{33} [30]. Qualification: A conventional MPB has not been established, and proposed MPB-like behavior remains to be verified.
Defect engineering	Defects were known to affect electrical properties, but their roles in polarization and domain behavior were unclear.	Confirmation: Nonstoichiometry studies reaffirm the influence of defects on electrical properties. Extension: A-site deficiency improves densification, breakdown strength, resistivity, and d_{33} , while B-site deficiency can introduce defect dipoles and domain-wall pinning [23]. Qualification: Greater structural distortion does not necessarily yield higher d_{33} ; intrinsic polarization and domain-wall mobility must be balanced.
Processing and densification	Poor densification was a major limitation; sintering aids and pressure-assisted sintering had already been explored.	Confirmation: Powder and sintering optimization improve densification. Extension: Mechanochemical activation and precursor control also regulate microstructure and densification [24,31–35]. Qualification: Higher relative density alone does not guarantee higher d_{33} ; defects, grain morphology, orientation, and poling are also critical.
Texturing Techniques	Preferred grain orientation was recognized as a promising route for exploiting the strong crystallographic anisotropy of PLS ceramics.	Confirmation: Preferred orientation enhances piezoelectric response. Extension: Recent textured Sr ₂ Nb ₂ O ₇ studies extend performance evaluation to thermal stability and elevated-temperature resistivity [36,37]. Qualification: Texture-processing routes require further optimization to better control grain orientation and simultaneously improve poling efficiency, d_{33} , and high-temperature electrical resistivity.

2. Crystal Structure and Origin of Spontaneous Polarization

2.1. Crystal Structure

PLS compounds are layered oxides derived from the ABO_3 perovskite structure by periodically interrupting the corner-sharing BO_6 octahedral network, with a general formula of $A_nB_nO_{3n+2}$ (or $ABO_{3+2/n}$) [38–40]. The A sites are typically occupied by large cations such as Sr^{2+} , Ca^{2+} , and La^{3+} , whereas the B sites are mainly occupied by Nb^{5+} , Ta^{5+} , and Ti^{4+} . Here, n denotes the number or average number of BO_6 octahedral layers within each perovskite slab. As shown in Figure 1, increasing n increases the slab thickness and gradually approaches the three-dimensional ABO_3 perovskite structure.

Different stacking sequences can occur at the same average n . For example, $n = 3(I)$ consists of uniform three-layer slabs, whereas $n = 3(II)$ is formed by alternating two- and four-layer slabs; similarly, $n = 4.5$ represents alternating four- and five-layer slabs. These stacking characteristics, together with BO_6 octahedral tilting and distortion, determine the crystallographic symmetry. Uniform odd-layer members are generally centrosymmetric, whereas several even-layer and mixed-layer members can adopt noncentrosymmetric structures [41,42]. Therefore, PLS ferroelectric studies have mainly focused on noncentrosymmetric members, particularly the $n = 4$ family.

The $n = 4$ $A_2B_2O_7$ compounds are the most extensively studied PLS ceramics. Their ferroelectric phases mainly include orthorhombic structures, represented by $Sr_2Nb_2O_7$ and $Sr_2Ta_2O_7$, and monoclinic structures, represented by $Ca_2Nb_2O_7$, $La_2Ti_2O_7$, $Nd_2Ti_2O_7$, and $Pr_2Ti_2O_7$ [41–45]. Among them, $Sr_2Nb_2O_7$ and $La_2Ti_2O_7$ have received particular attention because of their ultrahigh T_C values.

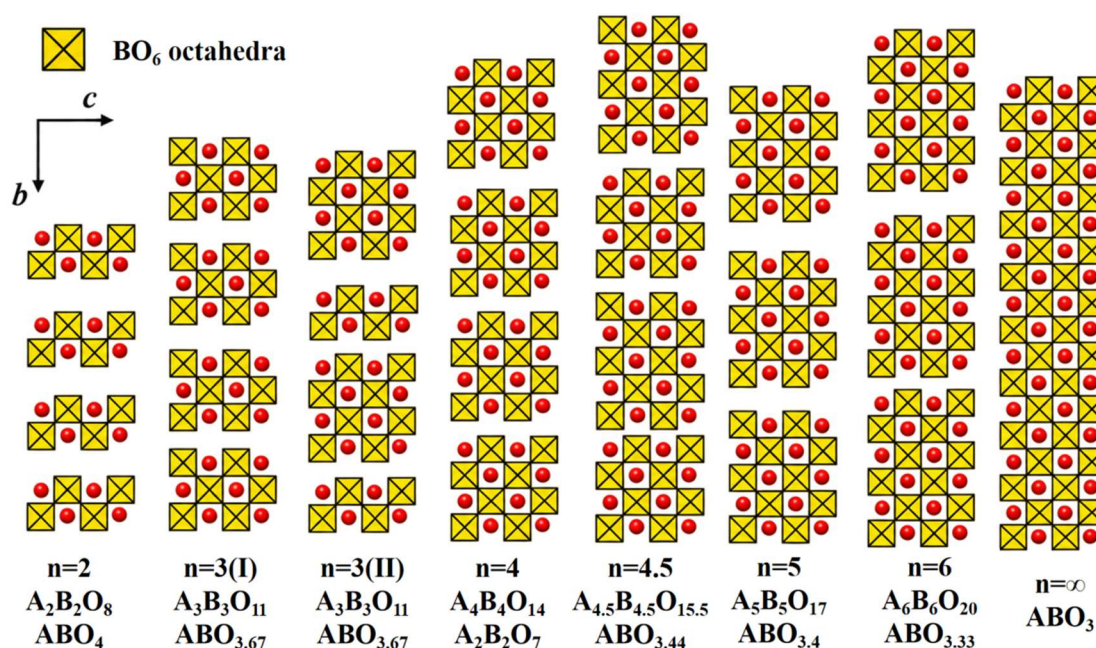


Figure 1. Schematic structures of PLS compounds with different n values [original figure prepared by the authors].

2.2. Origin of Spontaneous Polarization

The origin of spontaneous polarization is fundamental to understanding the ferroelectric phase transitions and piezoelectric responses of PLS ceramics. Owing to their unique layered structures, PLS compounds exhibit ferroelectric-to-paraelectric phase-transition pathways and origins of spontaneous polarization distinct from those of conventional perovskites, and are therefore regarded as typical oxygen-octahedra-driven ferroelectric materials [46]. Recent studies have substantially advanced the understanding of spontaneous polarization in PLS ferroelectrics.

Zhou et al. [25] investigated the ferroelectric-to-paraelectric phase-transition pathway and the origin of ferroelectricity in $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics. The results showed that the material undergoes a second-order incommensurate-to-commensurate phase transition at approximately 215 °C and a first-order ferroelectric-to-paraelectric phase transition at approximately 1340 °C. The high-temperature ferroelectric phase transition is primarily driven by the in-phase $a^0b^0c^+$ rotation of the NbO_6 octahedra about the c axis. As shown in Figure 2, soft-mode analysis revealed that the displacements of O, Sr, and Nb ions contribute 83.4%, 14.3%, and 2.2%, respectively, indicating that the spontaneous polarization of $\text{Sr}_2\text{Nb}_2\text{O}_7$ originates predominantly from oxygen-octahedral rotations, with Sr^{2+} displacements playing a secondary role and Nb^{5+} making only a minor contribution.

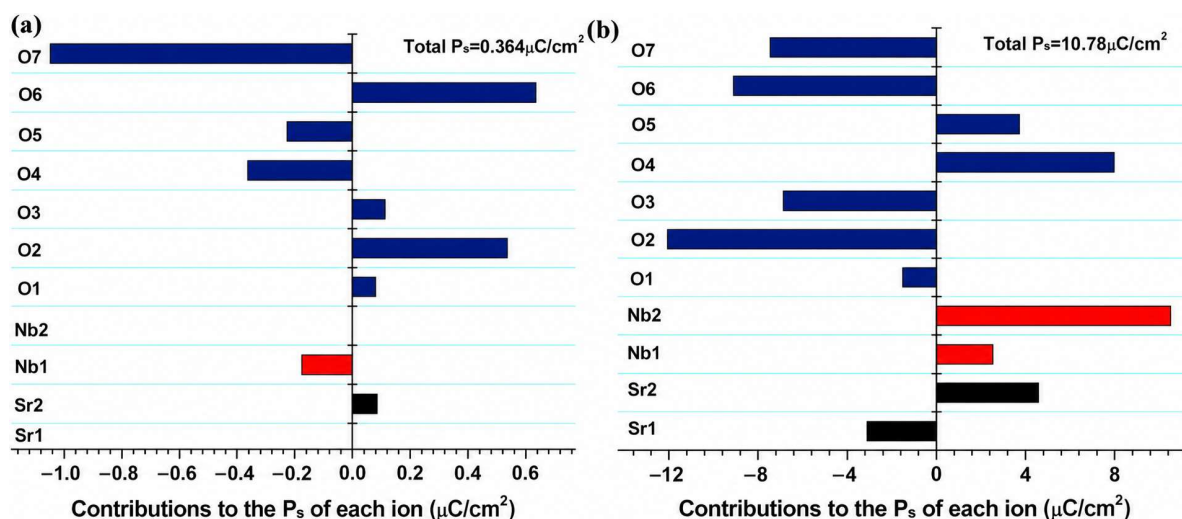


Figure 2. Contributions of individual atoms to the polarizability of $\text{Sr}_2\text{Nb}_2\text{O}_7$: (a) the incommensurate-to-commensurate phase transition at 215 °C; (b) the ferroelectric-to-paraelectric phase transition at 1340 °C [reproduced with permission from Elsevier [25], License No. 6336771245080].

Núñez Valdez et al. [26] employed first-principles calculations to investigate the origin and evolution of ferroelectricity in layered $\text{R}_2\text{Ti}_2\text{O}_7$ titanates ($\text{R} = \text{La}, \text{Ce}, \text{Pr}, \text{and Nd}$). The results showed that the ground state transforms from the centrosymmetric $P2_1/m$ phase to the polar $P2_1$ phase, with ferroelectricity primarily driven by a polar soft mode arising from the coupling of TiO_6 octahedral rotations and tilting with R^{3+} ion displacements along the b axis, while secondary nonpolar distortions further stabilize the ferroelectric ground state, indicating a typical intrinsic geometric ferroelectric mechanism. As the rare-earth ionic radius decreases from La to Nd, the phase-transition energy barrier and the degree of structural distortion progressively increase, whereas the spontaneous polarization remains nearly constant at approximately 18 $\mu\text{C}/\text{cm}^2$.

More recently, Zhang et al. [27] further demonstrated that out-of-plane tensile strain can enhance the in-plane polarization of $\text{La}_2\text{Ti}_2\text{O}_7$ by modifying oxygen-octahedral distortion and La-ion displacement, providing direct evidence that polarization in PLS materials can be tuned through local structural distortion.

Wang et al. [42] further revealed a pronounced orientation-dependent and thermally enhanced piezoelectric response in $\text{Ca}_2\text{Nb}_2\text{O}_7$ single crystals. Along the spontaneous-polarization b-axis, d_{33} increases from 10.4 pC/N at 273 K to 15.2 pC/N at 750 K. The enhancement was attributed to increased Ca^{2+} thermal motion and NbO_6 octahedral distortion at elevated temperatures, indicating that the coupled displacement of interlayer cations and oxygen octahedra plays an important role in the piezoelectric response. This thermally enhanced behavior, together with the ultrahigh T_C (~1773 K), highlights $\text{Ca}_2\text{Nb}_2\text{O}_7$ as a promising PLS material for high-temperature piezoelectric applications.

Overall, spontaneous polarization in PLS materials mainly arises from oxygen-octahedral rotations and tilting coupled with cation displacements. However, their quantitative contributions to domain behavior

and piezoelectric response remain unclear, highlighting the need for combined *in situ* characterization and theoretical calculations to establish clearer structure-property relationships.

3. Compositional Design

The low d_{33} values of PLS ceramics are primarily attributed to their restricted polarization directions, high coercive fields, and hindered domain-wall motion. Compositional design is therefore widely used to improve piezoelectric performance while retaining ultrahigh Curie temperatures. Current strategies mainly include A-site and B-site substitution, secondary-component solid solution, and defect engineering to regulate BO_6 octahedral distortion, defect states, and polarization behavior. More recently, computational screening has begun to complement experimentally driven composition optimization.

3.1. Chemical Doping

Early studies demonstrated that conventional A- or B-site substitution can improve the piezoelectric response of PLS ceramics, although the resulting d_{33} values generally remained below approximately 4 pC/N. In recent years, compositional modification has increasingly shifted toward mechanism-guided and multi-element regulation.

Li et al. [28] demonstrated that V substitution in $\text{La}_2\text{Ti}_2\text{O}_7$ induces local lattice distortion and regulates oxygen vacancies, increasing d_{33} to 4.8 pC/N. Tang et al. [29] constructed local structural heterogeneity through Li/Ce co-doping, which reduced the energy barrier for polarization switching and facilitated domain reversal, increasing the room-temperature d_{33} of $\text{La}_2\text{Ti}_2\text{O}_7$ ceramics to 6.4 pC/N and further to 7.4 pC/N after annealing at 500 °C, while retaining an ultrahigh Curie temperature of 1408 °C, as shown in Figure 3. Jabeen et al. [22] further introduced Mn modification into Li/Ce-doped $\text{La}_2\text{Ti}_2\text{O}_7$, achieving a room-temperature d_{33} of approximately 9 pC/N and retaining approximately 8.5 pC/N when measured at room temperature after thermal treatment at 1200 °C, indicating resistance to thermal depoling. More recently, V/Mn co-doping increased the d_{33} of $\text{Sr}_2\text{Nb}_2\text{O}_7$ to approximately 3.5 pC/N, which remained stable after annealing at 800 °C, and proposed that the synergistic regulation of enhanced intrinsic polarization and extrinsic domain-wall contributions is an effective strategy for improving the piezoelectric properties of $\text{Sr}_2\text{Nb}_2\text{O}_7$ [47].

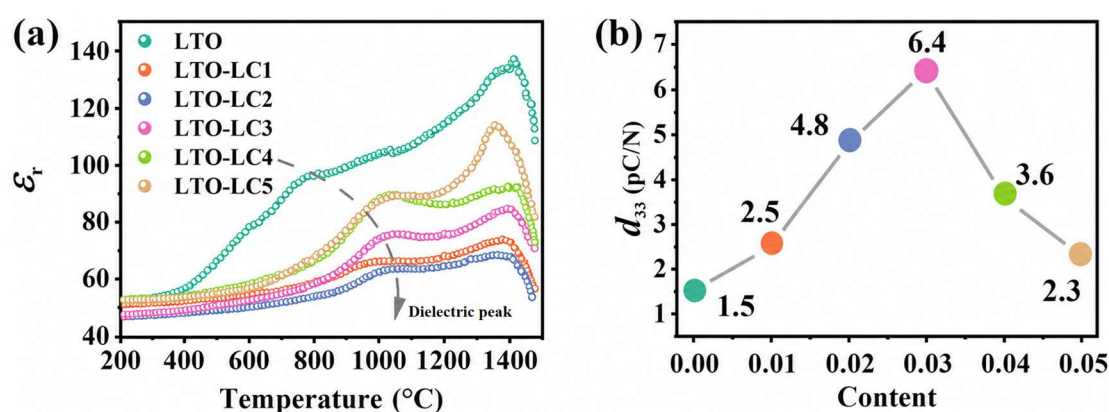


Figure 3. $\text{La}_{2-x}(\text{LiCe})_{x/2}\text{Ti}_2\text{O}_7$ ceramics: (a) temperature dependence of the relative permittivity at 1 MHz; (b) d_{33} values for different x values [reproduced with permission from the Royal Society of Chemistry [29], License No. 1774692-1].

Overall, chemical modification has increased the d_{33} of $\text{Sr}_2\text{Nb}_2\text{O}_7$ - and $\text{La}_2\text{Ti}_2\text{O}_7$ -based ceramics to approximately 3–4 and 9 pC/N [22], respectively. However, direct comparison of reported values should be treated with caution because differences in composition, densification, grain orientation, and poling conditions also affect the piezoelectric response. The available results suggest that multi-element

modification is particularly effective for $\text{La}_2\text{Ti}_2\text{O}_7$ by enhancing lattice distortion and facilitating polarization switching. Nevertheless, excessive doping may induce secondary phases, increase high-temperature leakage, or reduce T_C . Therefore, both piezoelectric enhancement and high-temperature stability should be considered in future compositional design.

3.2. Secondary-Component Solid Solution

Incorporating a secondary component to form a solid solution is an important compositional modification strategy for improving the sintering behavior and piezoelectric properties of PLS ceramics [30]. Unlike conventional perovskite piezoelectric ceramics, in which enhanced piezoelectricity is often associated with a morphotropic phase boundary (MPB), a well-defined MPB has not yet been established in PLS systems. Current solid solution studies mainly enhance d_{33} through lattice distortion, local structural heterogeneity, and improved poling conditions.

Early work demonstrated that introducing $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ into $\text{Sr}_2\text{Nb}_2\text{O}_7$ can promote densification while retaining $T_C > 1300^\circ\text{C}$, although the resulting d_{33} remained approximately 1.0 pC/N [48]. More recently, Sadhasivam et al. [30] investigated the isostructural $[\text{Sr}_2\text{Nb}_2\text{O}_7]_{1-x}[\text{La}_2\text{Ti}_2\text{O}_7]_x$ system and reported d_{33} values up to 9.3 pC/N. The enhancement was attributed to $\text{La}^{3+}/\text{Ti}^{4+}$ substitution at the Sr/Nb sites, resulting in $\text{NbO}_6/\text{TiO}_6$ octahedral distortions and enhanced local polarization. This recent result indicates that solid-solution design between structurally compatible PLS components may provide greater potential than conventional secondary-phase incorporation.

Overall, secondary-component solid solution enhances the d_{33} of PLS ceramics mainly through lattice distortion and local structural regulation. Although a conventional MPB has not yet been identified, MPB-like behavior may be achievable by coupling PLS end members with different structural symmetries or competing octahedral-rotation modes, provided that phase stability and high-temperature performance are preserved.

3.3. Defect Engineering

Defect engineering is an important strategy for tailoring the electrical properties of PLS ceramics. Unlike chemical doping and secondary-component solid solution, defect engineering primarily employs nonstoichiometric design to regulate vacancy concentrations, defect dipoles, and internal bias fields, thereby enhancing the dielectric breakdown strength and piezoelectric response of the ceramics.

Xia et al. [23] investigated the effects of A-site Sr deficiency on the structure and electrical properties of $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics. The results showed that an appropriate Sr deficiency increased the relative density from 92.1% to 96.8% and the dielectric breakdown strength from 206 kV/cm to 435–436 kV/cm, as shown in Figure 4, while increasing d_{33} from 1.0 to 2.1 pC/N and the electrical resistivity at 700°C from 6.7×10^5 to $1.1 \times 10^7 \Omega\cdot\text{cm}$. Raman analysis indicated that Sr deficiency improves the piezoelectric properties primarily by enhancing the tilting and rotation of NbO_6 octahedra, thereby increasing lattice distortion and the intrinsic polarization response. In contrast, B-site Nb deficiency produces more complex defect effects: it increases the c/a ratio and enhances lattice distortion, raising the d_{33} of $\text{Sr}_2\text{Nb}_{2-x}\text{O}_{7-2.5x}$ ceramics to approximately 1.4 pC/N; however, Nb vacancies readily associate with oxygen vacancies to form defect dipoles, resulting in pinched and shifted polarization-electric field hysteresis loops. As the measurement frequency increased from 1 Hz to 5000 Hz, the internal bias field increased from 1.1 to 4.5 kV/cm, indicating that the response of defect dipoles lags behind the applied electric field and constitutes an important cause of domain-wall pinning and restricted polarization.

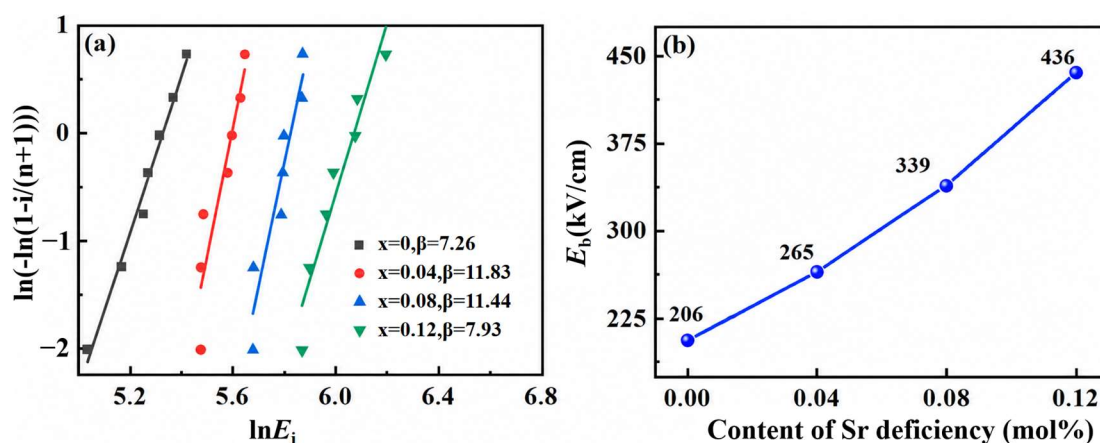


Figure 4. (a) Weibull distribution of dielectric breakdown strength for $\text{Sr}_{2-x}\text{Nb}_2\text{O}_{7-x}$ ceramics; (b) variation of dielectric breakdown strength with Sr deficiency content [reproduced with permission from Elsevier [23], License No. 6336201121187].

Overall, defect engineering has a dual effect on PLS ceramics, as increased structural distortion does not necessarily translate into an enhanced piezoelectric response. Therefore, defect control should balance enhancing intrinsic polarization with preserving domain-wall mobility.

Beyond experimentally driven compositional design, computational approaches have begun to provide guidance for identifying promising PLS compositions. In contrast to first-principles calculations primarily used to elucidate the microscopic origin of spontaneous polarization and structure-property relationships, computational screening aims to explore a broader compositional space and identify candidate materials for experimental validation. Ling et al. [49] employed a high-throughput computational strategy to screen 19 cation substitutions in $\text{Sr}_2\text{Nb}_2\text{O}_7$ and identified V-alloyed $\text{Sr}_2\text{Nb}_2\text{O}_7$ as a promising system. Large-cell DFT calculations predicted enhanced piezoelectricity, whereas thin-film experiments demonstrated only dielectric enhancement, leaving the piezoelectric prediction unverified. The initial screening uses approximate interpolation of end-member properties and requires further computational assessment. Direct piezoelectric measurements and validation in bulk ceramics under high-temperature conditions are therefore needed. Overall, computational screening of PLS materials remains in its early stages, and closer integration of theoretical predictions with experimental validation, particularly under high-temperature conditions, is needed to accelerate composition discovery.

4. Fabrication Processes

The fabrication process directly determines the powder reactivity and sintered density of PLS ceramics and is therefore a critical factor affecting their piezoelectric properties. Owing to the strong crystallographic anisotropy and poor densification of PLS ceramics during conventional sintering, recent studies have focused primarily on powder synthesis, low-temperature densification, advanced sintering techniques, and texturing processes to optimize the microstructure and enhance d_{33} .

4.1. Powder Synthesis

Early studies established solid-state reaction, molten-salt synthesis, and solution-based routes to control phase formation and particle morphology in PLS powders [50,51]. Recent research has shifted toward reducing synthesis temperature and tailoring precursor reactivity, particle size, and defect states to improve subsequent densification and piezoelectric performance. A mechanochemical route reported in 2019 reduced the temperature required for single-phase $\text{La}_2\text{Ti}_2\text{O}_7$ formation from approximately 1150 to 850 °C [31]. Shichalin et al. [32] subsequently prepared nanoscale $\text{La}_2\text{Ti}_2\text{O}_7$ powders using a citrate sol-gel route and combined them with SPS to fabricate dense ceramics, demonstrating the potential of integrating chemical powder synthesis with rapid densification.

More recently, Tang et al. [33] showed that the crystal phase and particle size of the TiO_2 precursor strongly affect the phase formation, microstructure, and electrical properties of $\text{La}_2\text{Ti}_2\text{O}_7$ ceramics, with nano-rutile TiO_2 providing superior overall performance. Li et al. [34] further demonstrated that mechanochemical activation can modify grain/domain structure and oxygen-vacancy states, producing $\text{La}_2\text{Ti}_2\text{O}_7$ ceramics with $d_{33} \sim 4.6$ pC/N and a resistivity of $2.1 \times 10^7 \Omega \cdot \text{cm}$ at 800°C . These results indicate that modern powder processing is evolving from simple phase synthesis toward coordinated control of powder reactivity, defects, microstructure, and high-temperature electrical properties.

4.2. Conventional Solid-State Sintering

The conventional solid-state reaction (SSR) method is simple and cost-effective, making it suitable for compositional modification studies; however, because PLS ceramic grains are predominantly plate-like and exhibit strong structural anisotropy, their random packing during conventional sintering readily generates pores, resulting in poor densification, low poling efficiency, and a relatively low d_{33} .

To address the low sintered density and weak piezoelectric response of conventionally sintered $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics, Chen et al. [24] investigated the effect of grain size on their piezoelectric properties, as shown in Figure 5. The results showed that $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics with average grain sizes of $1.2\text{--}3.6 \mu\text{m}$ could be obtained by adjusting the sintering temperature; as the grain size increased, the relative density rose to approximately 96%, and d_{33} increased to 1.7 ± 0.1 pC/N, while a high T_C of approximately 1340°C and a thermal depoling temperature above 1200°C were retained.

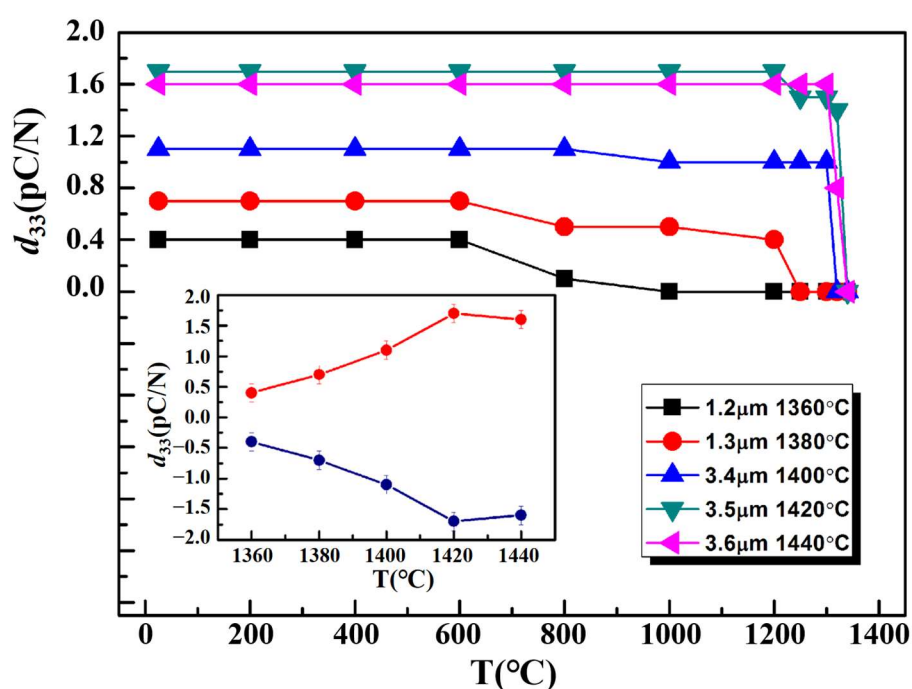


Figure 5. Temperature dependence of d_{33} values with different grain sizes after thermal annealing. Inset: d_{33} values of samples sintered at different temperatures; positive and negative values correspond to opposite sample clamping orientations during measurement [reproduced from World Scientific [24] under the applicable open-access license].

$\text{La}_2\text{Ti}_2\text{O}_7$ ceramics can also be fabricated via SSR, although relatively high sintering temperatures are generally required. Previous studies have shown that relatively dense $\text{La}_2\text{Ti}_2\text{O}_7$ ceramics can be obtained by sintering at $1400\text{--}1500^\circ\text{C}$, with d_{33} values of approximately $1.0\text{--}1.5$ pC/N; although further increasing the sintering temperature promotes grain growth, it may also induce residual strain or abnormal grain growth, thereby compromising the stability of the ferroelectric and piezoelectric properties [33].

Overall, SSR offers the advantage of a well-established process and serves as the foundation for both fundamental research and large-scale fabrication of PLS ceramics. However, conventional sintering alone cannot readily achieve high density, effective poling, and a high d_{33} simultaneously. Further studies, therefore, generally combine compositional design, sintering aids, and other strategies to improve densification and piezoelectric properties.

4.3. Low-Temperature Densification Sintering

Low-temperature densification sintering typically employs low-melting-point oxides or liquid-phase sintering aids to promote particle rearrangement and grain-boundary diffusion, thereby reducing the sintering temperature, increasing the density, and improving the subsequent poling conditions and piezoelectric response.

Earlier CuO-assisted sintering established that liquid-phase additives can substantially reduce the sintering temperature and improve the densification of $\text{Sr}_2\text{Nb}_2\text{O}_7$ [12]. More recent studies have extended this strategy to $\text{La}_2\text{Ti}_2\text{O}_7$ with substantially greater enhancement of piezoelectric performance. Tang et al. [52] introduced V_2O_5 as a sintering aid, reducing the sintering temperature from 1500 to 1400 °C and increasing d_{33} from approximately 1.5 to 5.6 pC/N while retaining $T_C > 1370$ °C. The improvement was associated with enhanced densification and local structural distortion. Swami et al. [35] subsequently showed that Bi_2O_3 -assisted sintering promotes grain growth and reduces porosity, increasing d_{33} from 1.4 to 2.1 pC/N. However, excessive Bi_2O_3 can readily cause volatilization, grain-boundary segregation, or secondary-phase precipitation; therefore, its addition level and the sintering schedule must be carefully controlled.

Notably, higher density does not necessarily translate into a higher d_{33} . CuO-assisted $\text{Sr}_2\text{Nb}_2\text{O}_7$ with ~98% relative density exhibited a d_{33} of 1.1 ± 0.1 pC/N, whereas conventionally sintered ceramics with ~96% density reached 1.7 ± 0.1 pC/N, indicating that the piezoelectric response is also strongly influenced by defect chemistry, grain morphology, crystallographic orientation, and poling efficiency.

Overall, low-temperature densification sintering can effectively lower the sintering temperature, improve densification, and facilitate poling of PLS ceramics; however, phase purity, high-temperature electrical resistivity, and piezoelectric stability must be carefully balanced to minimize sintering-aid volatilization, grain-boundary segregation, and high-temperature leakage.

4.4. Advanced Sintering Techniques

Conventional solid-state sintering relies primarily on thermal activation to achieve densification, whereas advanced sintering techniques introduce external pressure, electric current fields, or high-pressure environments to further improve the thermodynamic and kinetic conditions of sintering, providing an important processing route for overcoming the poor densifiability of PLS ceramics. The advanced sintering techniques widely reported to date mainly include spark plasma sintering (SPS), hot-press sintering (HP), hot-forging sintering (HF), flash sintering (FS), and cold sintering processing (CSP).

Early studies demonstrated that SPS [53], HP [54], and HF [55] can effectively improve the densification and crystallographic orientation of PLS ceramics, establishing the feasibility of pressure-assisted sintering for these highly anisotropic materials. However, most systematic investigations of these processes were conducted before 2018. Recent research on advanced sintering of PLS ceramics remains comparatively limited. Shichalin et al. [32] combined nanoscale sol-gel-derived $\text{La}_2\text{Ti}_2\text{O}_7$ powders with SPS to obtain dense ceramics over a relatively broad sintering temperature range, demonstrating the potential of integrating chemically synthesized nanopowders with rapid field-assisted densification. Nevertheless, systematic correlations among SPS parameters, texture, defect states, high-temperature resistivity, and d_{33} remain scarce. This represents an important gap for future processing studies.

4.5. Texturing Techniques

Ceramic texturing promotes the preferential alignment of grains along specific crystallographic directions, thereby producing anisotropic properties approaching those of single crystals along the preferred orientation. Texturing remains one of the most effective processing strategies for overcoming the restricted polarization direction of PLS ceramics.

Shi et al. [37] synthesized $\text{Sr}_2\text{Nb}_2\text{O}_7$ template grains using $\text{Na}_2\text{SO}_4/\text{K}_2\text{SO}_4$ molten salts and mixed them with $\text{Sr}_2\text{Nb}_2\text{O}_7$ matrix powder to prepare a tape-casting slurry, followed by tape casting, cutting, and sintering to obtain textured $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics. The resulting ceramics exhibited pronounced preferred orientation, with an orientation factor of $f = 0.78$, a d_{33} of 2.8 pC/N in the perpendicular direction, a T_C of 1328 °C, and an electrical resistivity of $5.6 \times 10^6 \Omega \cdot \text{cm}$ at 650 °C. More recently, Shao et al. [36] achieved an orientation factor of 0.86 in textured $\text{Sr}_2\text{Nb}_2\text{O}_7$ by regulating the microstructure, thereby increasing d_{33} to 3.2 pC/N. As shown in Figure 6, plate-like particles prepared by the molten-salt route promote preferential grain alignment during SPS, producing a textured microstructure, whereas nonagglomerated particles tend to form randomly oriented equiaxed grains. The textured ceramics retained a stable piezoelectric response up to approximately 1300 °C and an electrical resistivity of $1.33 \times 10^6 \Omega \cdot \text{cm}$ at 800 °C, demonstrating the effectiveness of texture engineering for ultrahigh-temperature piezoelectric applications.

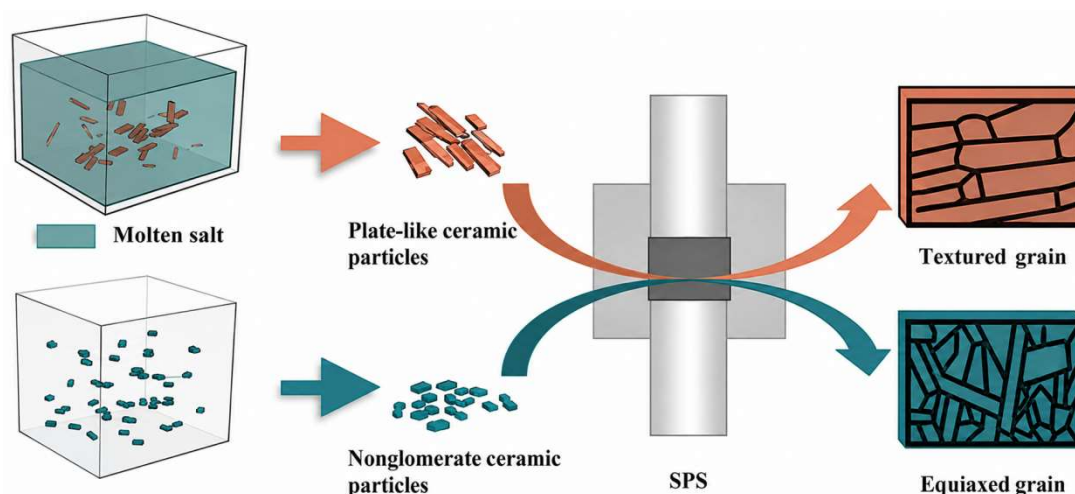


Figure 6. Schematic illustration of the preparation of $\text{Sr}_2\text{Nb}_2\text{O}_7$ ceramics with isotropic and textured microstructures [reproduced with permission from Wiley [36], License No. 6341100191425].

Earlier work combining strong-magnetic-field alignment with TGG established multidimensional texture control in $\text{La}_2\text{Ti}_2\text{O}_7$ [56]. Future studies should further optimize texture development and systematically correlate grain orientation with poling efficiency, d_{33} , and electrical resistivity at 600–1000 °C to identify processing conditions suitable for ultrahigh-temperature sensing.

Table 3 summarizes the distributions of d_{33} and T_C for PLS piezoelectric ceramics prepared using different methods in recent years. Most systems exhibit T_C values above 1200 °C, whereas their d_{33} values remain generally low, with early studies reporting values predominantly in the range of 1–4 pC/N. In recent years, strategies including V_2O_5 -assisted sintering, Li/Ce co-doping, and Mn modification have increased the d_{33} of $\text{La}_2\text{Ti}_2\text{O}_7$ -based ceramics to 5–9 pC/N while retaining a high T_C of approximately 1400 °C. These results demonstrate that the synergistic integration of compositional regulation and process optimization is an important route for enhancing the piezoelectric properties of PLS ceramics.

Table 3. Key properties of PLS piezoelectric ceramics prepared by different methods [adapted and updated from our previous review [1], with additional data compiled from the primary literature cited in the Ref. column].

Year	Materials	Process	Relative Density (%)	T_C (°C)	d_{33} (pC/N)	Ref.
1992	$\text{Sr}_2(\text{Nb}_{0.5}\text{Ta}_{0.5})_2\text{O}_7$	HF	99	823	1.6	[55]
2009	$\text{La}_2\text{Ti}_2\text{O}_7$	Two-step SPS	/	1461	2.6	[53]
2010	$\text{Sr}_2\text{Nb}_2\text{O}_7$	SPS	/	1327	2.8	[57]
2013	$\text{Sr}_{2-x}\text{Ba}_x\text{Nb}_2\text{O}_7$	SPS	95	1175	3.6	[58]
2013	$\text{La}_{2-x}\text{Ce}_x\text{Ti}_2\text{O}_7$	SPS	95	1440	3.9	[59]
2013	$(\text{Sr}_{1-x}\text{Ce}_x)_2\text{Nb}_2\text{O}_7$	SPS	98	1327	1.5	[60]
2013	$\text{Pr}_2\text{Ti}_2\text{O}_7$	Two-step SPS	/	>1555	0.5	[61]
2016	$(\text{Sm}_x\text{La}_{1-x})_2\text{Ti}_2\text{O}_7$	SPS	/	1430	2.8	[62]
2016	$(\text{Bi}_x\text{La}_{1-x})_2\text{Ti}_2\text{O}_7$	SPS	95	1395	3.6	[63]
2017	$(1-x)\text{Sr}_2\text{Nb}_2\text{O}_7-x\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$	SSR	96.8	1330	1.0	[48]
2017	$\text{Sr}_2\text{Nb}_2\text{O}_7-0.5 \text{ wt}\% \text{ CuO}$	SSR	98	1342	1.1	[12]
2017	$\text{Sr}_2\text{Nb}_2\text{O}_7$	HP	98	/	2.1	[54]
2018	$\text{Sr}_2\text{Nb}_2\text{O}_7$	SSR	96	1340	1.7	[24]
2020	$\text{La}_2\text{Ti}_2\text{O}_7$	SPS	/	>1400	4.7	[64]
2020	$\text{La}_2\text{Ti}_{1.96}\text{V}_{0.04}\text{O}_7$	SSR	/	/	4.8	[65]
2020	$\text{Sr}_2\text{Nb}_2\text{O}_7$	TGG	/	1328	2.8	[37]
2021	$\text{Nd}_2\text{Ti}_2\text{O}_7$	SSR	94.85	≥ 1500	2–3	[44]
2022	$\text{Sr}_{2-x}\text{Nb}_2\text{O}_{7-x}$	SSR	96.8	/	2.1	[23]
2023	$[\text{Sr}_2\text{Nb}_2\text{O}_7]_{1-x}[\text{La}_2\text{Ti}_2\text{O}_7]_x$	SSR	/	/	9.3	[30]
2024	$\text{V}_2\text{O}_5\text{-La}_2\text{Ti}_2\text{O}_7$	SSR	/	1370	5.6	[52]
2024	$\text{Li/Ce-La}_2\text{Ti}_2\text{O}_7$	SSR	/	1408	6.4	[29]
2024	$\text{La}_{1.97}(\text{LiCe})_{0.03}\text{Ti}_2\text{O}_7:0.2 \text{ wt}\% \text{ MnO}_2$	SSR	/	1415	9.0	[22]
2024	$\text{La}_{1.91}\text{Bi}_{0.09}\text{Ti}_{1.97}\text{V}_{0.03}\text{O}_7$	SSR	/	1437	4.0	[66]
2025	$\text{La}_{2-x}\text{Bi}_x\text{Ti}_2\text{O}_7$	SSR	/	1250	2.1	[35]
2025	$\text{Sr}_2\text{Nb}_2\text{O}_7$	Texture	/	1342	3.2	[36]
2025	$\text{La}_2\text{Ti}_2\text{O}_7$	SSR	/	>1400	4.6	[34]
2026	$\text{Sr}_2\text{Nb}_{2-x}\text{V}_x\text{O}_7-0.2\text{wt}\% \text{ MnO}_2$	SSR	/	1280	3.5	[47]

5. High-Temperature Performance and Stability

Although PLS ceramics are intended for operation above 600 °C, most reported d_{33} values are measured at room temperature. Direct characterization at 600–1000 °C remains limited because of the experimental challenges associated with ultrahigh-temperature piezoelectric measurements. Consequently, high-temperature performance is often evaluated indirectly through room-temperature measurements after thermal treatment, whereas *in situ* data are comparatively scarce. This limitation should be considered when comparing the thermal stability reported for different PLS systems.

Despite the limited high-temperature data, several PLS systems exhibit promising thermal stability. Mn-modified Li/Ce- $\text{La}_2\text{Ti}_2\text{O}_7$ retains a d_{33} of approximately 8.5 pC/N when measured at room temperature after thermal treatment at 1200 °C [22], while textured $\text{Sr}_2\text{Nb}_2\text{O}_7$ shows little degradation in piezoelectric response after treatment up to approximately 1300 °C and maintains an electrical resistivity of $1.33 \times 10^6 \Omega \cdot \text{cm}$ at 800 °C [36]. High-temperature insulation can also be substantially improved through defect and processing control, with Sr-deficient $\text{Sr}_2\text{Nb}_2\text{O}_7$ reaching $1.1 \times 10^7 \Omega \cdot \text{cm}$ at 700 °C [23] and mechanochemically activated $\text{La}_2\text{Ti}_2\text{O}_7$ reaching $2.1 \times 10^7 \Omega \cdot \text{cm}$ at 800 °C [34]. These results indicate that the practical high-temperature capability of PLS ceramics depends not only on the thermal retention of the piezoelectric response but also on the suppression of thermally activated electrical conduction.

Overall, available results demonstrate the potential of PLS ceramics for operation above 600 °C, but systematic high-temperature data remain scarce. Standardized *in situ* measurements and long-term thermal

aging are therefore needed to establish reliable operating limits and enable meaningful comparison among different PLS systems.

6. Conclusions and Prospects

PLS piezoelectric ceramics combine ultrahigh T_C , high ρ , and excellent thermal stability, making them promising for sensing above 600 °C. Their spontaneous polarization originates predominantly from BO_6 octahedral rotations and tilting coupled with A-site cation displacements. However, the same highly anisotropic layered structure limits polarization switching, leading to high coercive fields, difficult poling, poor densifiability, and relatively low piezoelectric coefficients.

Recent advances, particularly since 2018, demonstrate that both compositional design and processing regulation can improve the piezoelectric performance of PLS ceramics. Doping, solid-solution design, and defect engineering regulate lattice distortion, defect states, and polarization switching, increasing d_{33} to approximately 3–4 pC/N in $\text{Sr}_2\text{Nb}_2\text{O}_7$ -based ceramics and approximately 9 pC/N in $\text{La}_2\text{Ti}_2\text{O}_7$ -based systems while largely retaining their ultrahigh T_C . Meanwhile, powder activation, low-temperature densification, advanced sintering, and texture engineering improve powder reactivity, ceramic density, grain orientation, and high-temperature insulation. Nevertheless, increased density or crystallographic orientation does not necessarily yield a proportional increase in d_{33} , as the piezoelectric response is also governed by defect chemistry, grain morphology, domain mobility, and poling conditions. PLS ceramics should therefore be evaluated using multiple parameters, including d_{33} , T_C , high-temperature resistivity, dielectric breakdown strength, and thermal stability.

Despite these advances, substantial challenges remain. The d_{33} values of PLS ceramics are still considerably lower than those of PZT and most bismuth-layered piezoelectric ceramics, while high coercive fields and thermally activated electrical conduction limit effective poling and high-temperature operation. Moreover, most reported d_{33} values are measured at room temperature, and thermal stability is commonly assessed indirectly after heat treatment. Systematic *in situ* measurements at 600–1000 °C, long-term thermal-aging studies, and device-level validation remain scarce. Advanced sintering and texturing techniques also face limitations related to equipment dependence, processing cost, and sample dimensions.

Future research should focus on the following directions:

- (1) Elucidating the structural origin of piezoelectricity. The relationships among oxygen-octahedral rotation modes, local lattice distortion, domain structures, and temperature-dependent polarization should be clarified through *in situ* structural analysis, domain-scale characterization, and first-principles calculations.
- (2) Promoting synergistic composition-processing optimization. Compositional design should be integrated with powder synthesis, densification, texture control, and poling to simultaneously improve d_{33} , electrical resistivity, and dielectric breakdown strength without compromising phase purity or thermal stability.
- (3) Strengthening device-level validation. Further efforts are needed to develop high-temperature electrodes and packaging and to conduct long-term sensor testing above 600 °C, thereby promoting the transition of PLS ceramics from material development to practical sensing applications.

7. Review Methodology

The literature reviewed covers studies published from 1971 to 2026, with emphasis on advances since 2018. Publications were identified mainly through Web of Science and Google Scholar using keywords including “perovskite layer structure”, “PLS piezoceramics”, “high-temperature piezoelectric ceramics”, “ $\text{Sr}_2\text{Nb}_2\text{O}_7$ ” and “ $\text{La}_2\text{Ti}_2\text{O}_7$ ”. The literature selection focused on peer-reviewed studies addressing crystal structure and polarization mechanisms, compositional and defect engineering, fabrication and texturing processes, and high-temperature piezoelectric and electrical properties of PLS materials. Representative

studies were selected based on their relevance to these topics, availability of quantitative property data, mechanistic significance, and contribution to recent advances.

Statement of the Use of Generative AI and AI-Assisted Technologies in the Writing Process

During the preparation of this manuscript, the authors used DeepSeek to assist in translating the original Chinese data into English and proofreading the English manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

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Author Contributions

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Not applicable.

Informed Consent Statement

Not applicable.

Data Availability Statement

Data availability is not applicable to this article as no new data were created or analyzed in this study.

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Declaration of Competing Interest

The authors declare no competing financial interest.

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