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REVIEW

Brief Review of Lead-Free Piezoelectric Ceramics for High-Power Applications

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ABSTRACT: Lead-free piezoelectric ceramics have gained significant attention as environmentally friendly alternatives to lead-based materials such as $Pb(Zr,Ti)O_3$ (PZT) for high-power applications. This review focuses on three prominent lead-free ceramic systems: $BaTiO_3$ (BT), $Bi_{0.5}Na_{0.5}TiO_3$ (BNT), and $(Na_{0.5}K_{0.5})NbO_3$ (NKN). BT-based ceramics offer excellent piezoelectric properties but are limited by their low Curie temperatures, which has been addressed through doping and sintering aids such as Li_2O. BNT-based ceramics exhibit high thermal stabilities and tunable electromechanical properties; however, challenges remain because of their relatively low piezoelectric coefficients. NKN-based ceramics demonstrate outstanding mechanical quality factors (Q_m) and high Curie temperatures, with significant performance improvements achieved through CuO doping and co-doping with other oxides. Despite these advancements, issues such as thermal stability, alkaline element volatility, and limited d_{33} still need to be resolved. Continued progress in doping strategies, microstructural control, and advanced processing techniques will be crucial to fully unlock the potential of lead-free piezoelectric ceramics for high-power applications.

KEYWORDS: *Lead-free piezoelectric ceramics, High-power applications, Mechanical quality factor (Q_m), Doping strategies*

1. INTRODUCTION

Piezoelectric materials play a pivotal role in modern electronics and industrial technologies, especially in high-power applications [1-3]. Devices such as ultrasonic transducers, piezoelectric transformers, and sonar systems demand high precision and energy efficiency [4-6]. Piezoelectric materials, which convert electrical energy into mechanical energy and vice versa, are the core components of high-performance devices. The stability and reliability of piezoelectric materials are critical for high-power applications.

Piezoelectric ceramics are broadly categorized into two main types: "soft ceramics" and "hard ceramics." Soft piezoelectric ceramics are characterized by high piezoelectric coefficients (d_{33}) and sensitivity, making them suitable for sensors and low-power applications. By contrast, hard piezoelectric ceramics exhibit high mechanical quality factors (Q_m) and low dielectric loss, which are essential for high-power applications [7].

High-power piezoelectric applications often require a combination of hard and soft properties. For high-power applications, achieving a balance between the mechanical stability and low energy loss of hard ceramics and the sensitivity and high energy-conversion efficiency of soft ceramics is critical. The harmonious integration of these properties is not only vital for efficient operation but also for ensuring stable and durable performance over time. Consequently, the development of piezoelectric materials that combine or optimize both characteristics remains a key challenge for high-power applications.

Environmental regulations and concerns about lead toxicity have incited research into lead-free piezoelectric ceramics as alternatives to lead-based materials, particularly $Pb(Zr,Ti)O_3$ (PZT) [8-10]. The European Union's Restriction of Hazardous Substances Directive has further emphasized the need to limit lead use in electronic products, accelerating the development of lead-free alternatives. Although PZT has been widely used owing to its exceptional electromechanical properties, environmental issues associated with lead have shifted

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attention to lead-free materials such as $BaTiO_3$ (BT), $(Bi_{0.5}Na_{0.5})TiO_3$ (BNT), and $(Na_{0.5}K_{0.5})NbO_3$ (NKN) [11-13].

Research on lead-free piezoelectric ceramics has focused on achieving eco-friendly materials with high mechanical quality factors and piezoelectric coefficients suitable for high-power applications. These properties can be further enhanced through advanced doping strategies and microstructural control [14-16]. Recent studies have highlighted the importance of texturing techniques that optimize the alignment of crystallographic grains to improve the electromechanical performance [17]. Texturing has emerged as a critical approach to enhancing the properties of lead-free hard ceramics, making it a key focus of contemporary research.

Current studies on lead-free piezoelectric ceramics primarily concern specific material systems such as BT, BNT, and NKN, each of which exhibits unique physical and chemical properties. Researchers have continued to improve their performance by employing various doping strategies and processing techniques. This review aims to provide a comprehensive analysis of lead-free hard piezoelectric ceramics and discuss their potential applications, existing limitations, and prospects for high-power devices. Finally, this study seeks to explore the commercial viability of lead-free piezoelectric ceramics for advanced technological applications.

2. BT-based High-power Piezoceramics

Barium titanate ($BaTiO_3$), the first perovskite-structured piezoelectric material, has historically played a vital role in applications such as underwater transducers [18]. Although its widespread use has decreased with the advent of PZT ceramics, BT remains a prominent contender among lead-free piezoelectric materials owing to its exceptional electromechanical characteristics.

Pure BT ceramics produced using conventional solid-state methods typically exhibit a room temperature piezoelectric coefficient (d_{33}) of approximately 180 pC/N. Efforts to enhance these properties have focused on alternative preparation techniques such as hydrothermal and microwave sintering methods [19-21]. However, these advanced techniques generally involve trade-offs such as reduced mechanical quality factors (Q_m), which may limit their suitability for specific applications.

The sintering process for BT-based ceramics generally requires temperatures above 1300°C to achieve sufficient material density. Researchers have investigated the use of sintering aids to address this issue. For example, the addition of 1 mol% Li_2O has demonstrated notable improvements in electromechanical properties, achieving a Q_m of ~1405 and a d_{33} of ~255 pC/N. These modifications underscore the

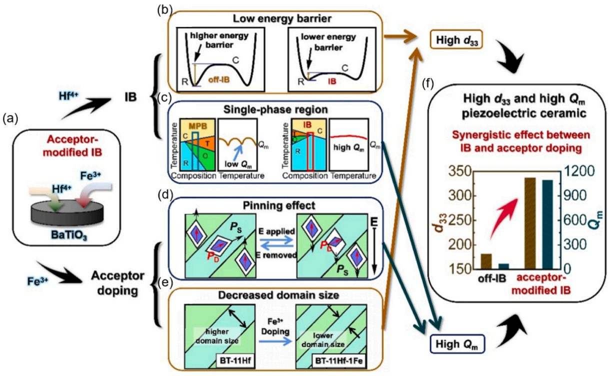

Fig. 1. Schematic of high Q_m and high d_{33} from the acceptor-modified IB. (a) Acceptor-modified IB was obtained in Hf^{4+} and Fe^{3+} doped BT ceramics. (b) IB *composition has a lower energy barrier than that of the off-IB* compositions, which leads to high d_{33}. (c) IB composition is a single-phase state and favorable to Q_m. (d) Acceptor doping generates a normal pinning effect, which is beneficial for improving Q_m. (e) Acceptor doping can decrease domain size, which is beneficial to improve d_{33}; (f) Piezoelectric ceramics with high d_{33} and high Q_m can be obtained using the synergistic effect of the acceptor-modified IB. [14] Copyright (2025) Elsevier

potential for tuning BT-based ceramics to meet specific application demands [11].

A recent study demonstrated that the tradeoff between the mechanical quality factor and piezoelectric coefficient in $BaTiO_3$-based ceramics can be addressed using acceptor-modified invisible boundary (IB) engineering, as shown in Fig. 1 [14]. By introducing Fe^{3+} dopants into $BaTi_{0.89}Hf_{0.11}O_3$ ceramics, researchers achieved a remarkable combination of Q_m ~1096 and d_{33} ~337 pC/N, outperforming many lead-free and lead-based piezoceramics. This approach leverages the low energy barrier for polarization rotation at the IB and pinning effect from acceptor doping, resulting in significant enhancements in electromechanical properties. However, the material suffers from a notably low Curie temperature (T_C) of only 48°C, which severely limits its applicability in high-temperature environments. These findings highlight the potential of IB engineering for developing high-performance lead-free piezoelectric materials while emphasizing the critical need for strategies to improve T_C for broader application viability.

One major limitation of BT-based ceramics is their relatively low Curie temperature (T_C), which is often below 100°C, restricting their application in high-temperature environments. Doping strategies have been effective in overcoming this challenge. For instance, MnO_2-doped BT ceramics have achieved a T_C of 123°C with a moderate Q_m of ~340.8. Similarly, $MnCO_3$ doping has produced BT ceramics with an ultrahigh Q_m of ~2400 and a T_C of ~133°C using traditional sintering techniques [22,23].

Table 1. Piezoelectric properties of BT-based, BNT-based, and NKN-based piezoelectric ceramics

Ceramics	Composition	Q_m	d_{33} (pC/N)	k_p	tan δ	ε_r	T_c (°C)	Refs.
BT-based	$(Ba_{0.99}Ca_{0.01})(Ti_{0.99}Co_{0.005}Nb_{0.005})O_3$ + 1 mol% Li_2O	1405	255	0.435	-	1862	111	[11]
	$(Ba_{0.97}Ca_{0.03})(Ti_{0.96}Sn_{0.035}Hf_{0.035})O_3$ + 2 mol% MnO_2	340	230	0.364	0.029	1289	123	[23]
	$(Ba_{0.95}Ca_{0.05})(Ti_{0.90}Sn_{0.10})O_3$ + 4 mol% Li_2O	491	457	0.251	0.023	12000	46	[16]
	$Ba(Ti_{0.89}Hf_{0.11})O_3$ + 1 mol% Fe_2O_3	1096	337	0.28	0.023	5000	48	[14]
BNT-based	$[Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}]TiO_3$ + 1.4 mol% MnO_2	470	142	0.289	0.029	570	320	[24]
	$(Bi_{0.485}Na_{0.425}K_{0.06}Ba_{0.03})TiO_3$ + 0.45 wt% MnO	380	170	0.295	0.017	790	250	[12]
	$[Bi_{0.5}(Na_{0.88}K_{0.08}Li_{0.04})_{0.5}](Ti_{0.985}Mn_{0.015})O_3$	974	85	0.218	0.009	308	305	[25]
	$0.88(Bi_{0.5}Na_{0.5})TiO_3$-$0.08(Bi_{0.5}K_{0.5})TiO_3$-$0.04BaTiO_3$	1200	105	0.193	0.01	400	265	[26]
	$0.94(Bi_{0.5}Na_{0.5})TiO_3$-$0.06BaTiO_3$ + 0.5 mol% MgO	826	112	0.29	-	-	-	[15]
NKN-based	$(Na_{0.5}K_{0.5})NbO_3$ + 1mol% MnO_2	370	102	0.38	-	350	414	[13]
	$(Na_{0.5}K_{0.5})NbO_3$ +1.5 mol% CuF_2	2708	91	0.36	0.003	250	420	[27]
	$(Na_{0.5}K_{0.5})(Nb_{0.996}Cu_{0.01})O_3$	3110	83	0.38	0.002	320	425	[28]
	$(Na_{0.52}K_{0.48})NbO_3$ + 1 mol% CuO + 1 mol% SnO_2	1040	120	0.38	0.013	705	378	[29]
	$(Na_{0.52}K_{0.47}Li_{0.01})NbO_3$ + 1 mol% CuO	1023	109	0.36	0.015	310	405	[30]
	$0.95(Na_{0.5}K_{0.5})NbO_3$-$0.05Li(Nb_{0.5}Sb_{0.5})O_3$ + 0.8 mol% CuO	320	207	0.44	0.014	340	-	[31]
	$[(Na_{0.5}K_{0.5})_{0.95}Li_{0.05}](Nb_{0.96}Sb_{0.04})O_3$ + 1.5 mol% CuO	560	170	0.37	-	600	-	[32]
	$(Na_{0.5}K_{0.5})NbO_3$ + 0.38 mol% $K_{5.4}Cu_{1.3}Ta_{10}O_{29}$ + 0.25 mol% MnO_2	1900	90	0.4	0.003	300	404	[33]
	$(Na_{0.5}K_{0.5})NbO_3$ + 0.38 mol% $K_{5.4}Cu_{1.3}Ta_{10}O_{29}$ + 0.5 mol% CuO	3053	94	0.38	0.002	285	-	[34]
	$(Na_{0.5}K_{0.5})(Nb_{0.9}Ta_{0.1})O_3$ + 1 mol% $K_4CuNb_8O_{23}$ + 0.5 mol% MnO_2	1563	96	0.422	0.004	308	340	[35]
	$[(Na_{0.535}K_{0.485})_{0.93}Li_{0.07}](Nb_{0.942}Ta_{0.058})O_3$ + 1 mol% CuO	323	185	0.37	0.008	500	422	[36]
	$0.97[(Na_{0.52}K_{0.48})_{0.82}Ba_{0.02}Ca_{0.04}Li_{0.02}]Nb_{0.85}O_3$-$0.03K_{0.85}Ti_{0.85}Nb_{1.15}O_5$ + 2 mol% MnO_2	650	140	0.42	0.005	750	320	[37]
	$0.965(Na_{0.55}K_{0.45})NbO_3$-$0.035Bi_{0.5}Na_{0.5}ZrO_3$ + 1 mol% CuO + 0.2 mol% Fe_2O_3	355	231	0.44	0.02	800	366	[38]
	$(Na_{0.5}K_{0.5})NbO_3$ + 0.75 mol% CuO + 3wt% $NaNbO_3$ templates	582	134	0.58	0.003	350	415	[17]

Increasing the T_C of BT-based ceramics remains a critical objective for their use in high-power applications. Advancements in doping and sintering methodologies hold promise for enhancing both the thermal and electromechanical properties of BT-based systems, enabling their broader adoption in demanding industrial and technological applications. The piezoelectric properties of BT, BNT, and NKN are summarized in Table 1.

3. BNT-based High-power Piezoceramics

Bismuth sodium titanate ($Bi_{0.5}Na_{0.5}TiO_3$; BNT) was first identified by Smolensky et al. in 1960 [39]. At room temperature, BNT exhibits a rhombohedral symmetry (R phase) and a relatively high Curie temperature (T_C) of approximately 320°C [40]. Given its potential for high-power applications, extensive efforts have been made to improve the electromechanical properties of BNT-based ceramics through doping strategies and optimized sintering processes to enhance their mechanical quali ty factor (Q_m).

Pure BNT ceramics were reported to achieve a moderate Q_m of ~366 and a low dielectric constant (ε_r) of ~345 at room temperature [41]. Structural and piezoelectric studies on $Bi_{0.5}(Na_{1-x}Li_x)_{0.5}TiO_3$ compositions indicated that doping with Li can slightly enhance Q_m, with a maximum increase of ~30 observed at x=0.04 [42]. However, excessive Li doping leads to deteriorated properties, underscoring the delicate balance required in compositional modifications. MnO and MnO_2 doping have also shown significant improvements in the electromechanical performance of BNT-based ceramics [12,24]. For instance, introducing up to 1.5 mol% Mn into $Bi_{0.5}(Na_{0.88}K_{0.08}Li_{0.04})_{0.5}(Ti_{1-x}Mn_x)O_3$ compositions increased Q_m from 402 to 974, albeit with a slight reduction in the piezoelectric coefficient (d_{33}) and electromechanical coupling factor (k_p) [43].

Innovative sintering techniques, such as flash sintering, have also been explored for BNT-based ceramics. Although this method reduces the grain size and consequently decreases Q_m (from 980 to 650) compared to conventional sintering, it offers advantages in terms of processing efficiency and material densification [44].

Acceptor doping on both the A- and B-sites within the perovskite lattice has been explored to improve the performance. Notably, introducing bismuth deficiency in

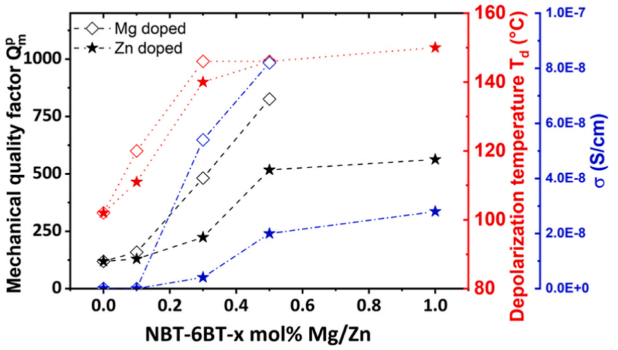

Fig. 2. Comparison of the mechanical quality factor Q_m (black dashed), depolarization temperature Td (red dotted), and conductivity σ (blue dashed/dotted) at RT between Mg-doped (rectangular) and Zn-doped (star) NBT-6BT with different amounts of doping concentration. [15] Copyright (2022) Elsevier

$0.88Bi_{0.50-x}Na_{0.50}TiO_3$-$0.08Bi_{0.50-x}K_{0.50}TiO_3$-$0.04BaTiO_3$ (BNKBT88) ceramics significantly enhances Q_m. While stoichiometric BNKBT88 ceramics have a Q_m of ~150, Bi-deficient ceramics achieve a remarkable Q_m of ~1200 owing to oxygen vacancies and domain wall pinning caused by intrinsic defects [26].

A recent study explored the effects of Mg doping on the $0.94Na_{1/2}Bi_{1/2}TiO_3$-$0.06BaTiO_3$ (NBT-6BT) lead-free piezoceramic system, revealing significant improvements in the mechanical quality factor and depolarization temperature (T_d) [15]. Mg doping increased the tetragonal phase fraction and non-cubic distortion, leading to a remarkable increase in Q_m to 826 in NBT-6BT-0.5Mg, as shown in Fig. 2. This high Q_m was stable under high vibration velocities, making the material suitable for high-power applications. However, piezoelectric coefficients decreased with increased doping, from 140 pC/N in undoped NBT-6BT to 112 pC/N in NBT-6BT-0.5Mg. These findings underscore the potential of Mg doping in enhancing the performance of NBT-BT ceramics for demanding applications while also highlighting the trade-offs in piezoelectric response.

Doping strategies and the introduction of secondary phases have been explored to enhance the performance of BNT-based ceramics. However, despite these efforts, BNT-based ceramics still face challenges in high-power applications owing to their relatively low piezoelectric coefficients (d_{33}) and electromechanical coupling factors (k_p). Addressing these limitations through advanced material design and processing techniques is essential to fully realize the potential of BNT-based ceramics for advanced high-power applications.

4. NKN-based High-power Piezoceramics

Potassium sodium niobate ($Na_{0.5}K_{0.5}NbO_3$, NKN) is a mixed lead-free piezoelectric system that combines orthorhombic potassium niobate ($KNbO_3$, KN) and sodium niobate ($NaNbO_3$, NN). Initially identified as a ferroelectric material by Shirane in 1954 [45], NKN exhibits an orthorhombic phase (Amm2) at RT. On heating, the phase structure transitions from orthorhombic (Amm2) to tetragonal (P4mm) at approximately 200°C and further to a cubic phase (m3m) above its Curie temperature (T_C) of approximately 420°C.

In 1959, Egerton and Dillon reported the piezoelectric and dielectric properties of pure NKN ceramics, which demonstrated a relatively low mechanical quality factor (Q_m) of ~130 and a modest piezoelectric coefficient (d_{33}) of ~80 pC/N [46]. Subsequent work by Du et al. showed that a relative density of 97.6% can be achieved using pressureless sintering, leading to improved properties with Q_m~210 and d_{33}~120 pC/N [47]. However, one of the key challenges in processing NKN-based ceramics is their poor sinterability, owing to the high volatility of alkaline elements at increased temperatures, making it difficult to achieve full densification and stoichiometric control.

To overcome these limitations, sintering aids have been widely used to enhance the sinterability and electromechanical performance of NKN ceramics. CuO, in particular, has proven effective in reducing sintering temperatures while improving Q_m and other performance metrics. In Li/Ta-modified NKN systems, CuO acts as an acceptor dopant that enhances sintering behavior and significantly increases Q_m [36]. Notably, Tan demonstrated that Cu^{2+}-doped NKN ceramics can achieve a Q_m as high as ~1241, whereas ultrahigh Q_m values exceeding 2500 have also been reported for Cu-doped NKN ceramics [48]. Interestingly, CuO addition imparts some “soft” characteristics, likely owing to the substitution of Cu^{2+} for Li^+, leading to a reduction in the coercive field (E_C) and Curie temperature (T_C).

In addition to CuO, codoping with other oxides has been explored as an effective strategy for enhancing the overall electromechanical performance of NKN ceramics [29,49,50], particularly for high-power applications. Small additions of compounds, such as $K_4CuNb_8O_{23}$ (KCN) and $K_{5.4}Cu_{1.3}Ta_{10}O_{29}$ (KCT), also improve the sinterability without compromising the piezoelectric properties [51]. For example, Matsubara et al. reported an increase in Q_m to 1200 when KCN was used as a sintering aid. Codoping with oxides in combination with KCN or KCT further enhances piezoelectric and dielectric properties, achieving significant improvements in Q_m [34].

A recent study on textured NKN ceramics highlighted an innovative approach that combined acceptor doping and texturing techniques to achieve high mechanical quality factors and piezoelectric coefficients, as shown in Fig. 3. By incorporating CuO as a dopant and $NaNbO_3$ templates for texturing,

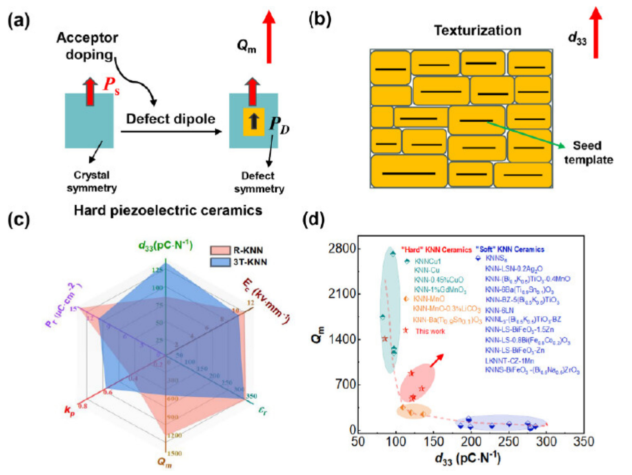

Fig. 3. NKN piezoelectric ceramics. Acceptor doping made the piezoelectric ceramics "hardened" and the Q_m value increased (a). The texture technique was used to improve d_{33} (b). A comparison of piezoelectric properties between R-NKN and 3T-NKN (c). The d_{33} and Q_m for different NKN-based piezoceramics, and with high Q_m and high d_{33} reported in paper (d). [17] Copyright (2024) American Chemical Society

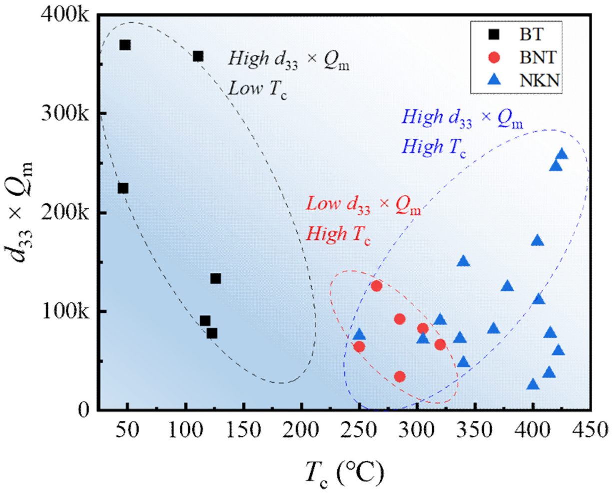

Fig. 4. d_{33}*Q_m versus Curie temperature (T_C) of BT-base, BNT-based, and NKN based piezoelectric ceramics

the researchers achieved exceptional properties, such as d_{33}~134, Q_m~582, k_p = 0.58, and a Curie temperature (T_C) of approximately 415°C. The texturing process enhanced grain orientation along the <001> direction, leading to improved piezoelectric performance while maintaining high mechanical robustness. This study demonstrates the potential of textured NKN ceramics as promising candidates for replacing lead-based piezoelectric materials in high-power applications. Further optimization of the processing conditions could enhance their performance for broader industrial use.

Although NKN-based ceramics exhibit substantial potential for high-power applications owing to their ultrahigh Q_m and favorable electromechanical properties, challenges remain. The volatility and hygroscopic nature of alkaline elements, along with the relatively low dielectric constants and piezoelectric coefficients (d_{33}), hinder their full utilization. Future research must focus on addressing these issues to further optimize NKN-based ceramics, particularly by improving their thermal and chemical stability while enhancing d_{33} and the dielectric constant (ε_r) without sacrificing their high Q_m. The d_{33}*Q_m versus Curie temperature (T_C) of BT-, BNT-, and NKN-based piezoelectric ceramics are summarized in Fig. 4.

5. CONCLUSIONS

Lead-free piezoelectric ceramics, including BT, BNT, and NKN, have emerged as strong candidates for replacing lead-based materials, such as PZT, in high-power applications. Each system offers unique advantages, but challenges remain in achieving optimized performance for practical use. BT-based ceramics demonstrate excellent piezoelectric properties but suffer from a low Curie temperature (T_C). Enhancements through advanced sintering techniques and doping, such as Li_2O additions, have improved their mechanical quality factor (Q_m) and piezoelectric coefficients (d_{33}). However, increasing their thermal stability remains essential for broader applicability. BNT-based ceramics benefit from a high T_C (~320°C) and tunable electromechanical properties. Although doping strategies, such as Mn or Bi-deficiency, have significantly improved their Q_m, these materials still exhibit relatively low d_{33} and electromechanical coupling factors, which limits their use in high-power applications. NKN-based ceramics offer remarkable potential, particularly owing to their ultrahigh Q_m and high T_C. The addition of sintering aids such as CuO and co-doping with various oxides has improved their sinterability and performance. Nonetheless, challenges related to the volatility and hygroscopic nature of alkaline elements must be addressed. Overall, lead-free piezoelectric ceramics show promising advancements but require further optimization to overcome their respective limitations. Continued research into doping, microstructural control, and sintering processes will be key to fully realizing their potential for high-power applications in the industrial and technological sectors.

CRediT Authorship Contribution Statement

Dong-Gyu Lee: Investigation, Methodology, Writing – original draft. **Seung-Bum Kim:** Investigation, Methodology. **Yongke Yan:** Writing - review & editing. **Sunghoon Hur:** Writing - review & editing, Supervision, Funding acquisition. **Hyun-Cheol Song:** Writing - review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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