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High-performance KNN-based piezoelectric ceramics for buzzer application

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Piezoelectric ceramic materials are important components of piezoelectric buzzers, where the parameter of inverse piezoelectric coefficient (d_{33}^*) plays a decisive role in the performance of the buzzer. Here, we report the manufacture and performance of a lead-free ceramic-based $(0.96(K_{0.5}Na_{0.5})(Nb_{0.96}Sb_{0.04})O_3$–$0.04(Bi_{0.5}Na_{0.5})ZrO_3$–1 mol% Al_2O_3, abbreviated as KNNS-BNZ-1 mol% Al_2O_3) piezoelectric buzzer and compare it with commercial ($PbZr_{0.5}Ti_{0.5}O_3$, abbreviated as PZT) ceramics. Briefly, KNN-based ceramics have a typical perovskite structure and piezoelectric properties of $d_{33} = 480$ pC/N, $k_p = 0.62$ and $d_{33}^* = 830$ pm/V, compared to $d_{33} = 500$ pC/N, $k_p = 0.6$ and $d_{33}^* = 918$ pm/V of the commercial PZT-4 ceramics. Our results show that the KNNS-BNZ-1 mol% Al_2O_3 ceramics have a similar sound pressure level performance over the testing frequency range to commercial PZT ceramics (which is even better in the 3–4 kHz range). These findings highlight the great application potential of KNN-based piezoelectric ceramics.

Keywords: lead-free piezoelectric ceramics, phase structure engineering, buzzer, atomizer

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1. Introduction

A piezoelectric buzzer primarily consists of a multi-resonator oscillator, a piezo buzzer, an impedance matcher, a resonance chamber, housing, and so on. The piezo buzzer is made of piezoelectric ceramic material, functioning as the main part of the piezoelectric buzzer and one of the most widely used products in the field of piezoelectric ceramics. Nowadays, the market of the piezoelectric ceramic materials is mainly occupied by $PbZr_{0.5}Ti_{0.5}O_3$-based (PZT-based) ceramics, due to their outstanding electric field induced strain properties and fatigue resistance.[1–7] Given that lead is hazardous to the environment and human health, lead-free piezoelectric ceramic materials have become a new hot research field, including $Bi_{0.5}Na_{0.5}TiO_3$-based (BNT-based), $BaTiO_3$-based (BT-based), $K_{0.5}Na_{0.5}NbO_3$-based (KNN-based) and $BiFeO_3$-based (BF-based).[8–10] Furthermore, due to the successive restrictions on the use of lead in different countries and regions, research on lead-free piezoelectric ceramic materials has been promoted.[10,11]

Due to the advantages of high Curie temperature, low strain hysteresis and low driving electric field, KNN-based ceramics have become one of the most promising candidates for actuator applications.[12–14] A few strategies have been used to promote the ceramic properties, such as phase structure engineering, defect engineering, sintering technology and so on.[10,15–19] Wu *et al.* constructed a multi-phase (R–O–T) coexistent KNN-based ceramic, where the piezoelectric constant and temperature stability were elevated through the flattened free energy, which promoted the reversal of ferroelectric domains, resulting in outstanding piezoelectric properties ($d_{33} = 500$ pC/N, $k_p = 0.5$) and great practical application performance in an ultrasonic transducer.[20] In addition, the inverse piezoelectric constant is a crucial parameter in applications such as buzzers and atomizers, and an excellent inverse piezoelectric constant helps to broaden the application scenarios of piezoelectric ceramics. Guo *et al.* found that an exotic bipolar strain performance could be obtained in Sr-doped KNN ceramics by defect engineering and finally gained a remarkably high piezoelectric coefficient of 2100 pm·V^{-1}.[21] After decades of effort, the properties of KNN-based lead-free piezoelectric ceramics are comparable with commercial PZT-based ceramics.

With the increasing performance of KNN-based piezoelectric ceramics, a growing number of application experiments of KNN-based lead-free piezoelectric ceramics were reported. Chu *et al.* designed lead-free $(Na_{0.473}K_{0.473}Li_{0.04})Nb_{0.883}Ta_{0.05}Sb_{0.06}O_3$–$0.007SrTiO_3$ (NKLNTS-0.007ST) ceramics and piezoelectric acoustic actuators prepared by multi-layer technology and the sound pressure level (SPL) was around 60 dB.[22] Guo *et al.* prepared $0.955(K_{0.48}Na_{0.52})(Nb_{0.99}Sb_{0.01})O_3$–$0.045(Bi_{0.5}Na_{0.5})ZrO_3$–0.5 mol%$MnO_2$ piezoelectric ceramics with high Q_m (150) and d_{33} (370 pC/N). The ceramics were manufactured into an atomizer and the atomizer quality was > 0.6 mL/min.[23] All

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these application experiments indicated a wonderful future for the KNN-based piezoelectric ceramics.

KNN-based ceramics exhibit excellent piezoelectric and acoustic properties, making them a potential substitute for PZT ceramics in the fabrication of piezoelectric buzzers. On one hand, its maximum piezoelectric constant d_{33} is close to that of PZT ceramics, with a Curie temperature around 250 °C. On the other hand, it features lower power consumption, effectively reducing operational costs. Therefore, using KNN-based ceramics for piezoelectric buzzers holds significant commercial potential.

In this paper, we focus on the practical application of ceramic materials in buzzers. To meet the core demand for high inverse piezoelectric performance, $0.96(K_{0.5}Na_{0.5})(Nb_{.96}Sb_{0.04})O_3$–$0.04(Bi_{0.5}Na_{0.5})ZrO_3$ ceramics were selected for compositional optimization, with the addition of Al_2O_3 as a sintering aid to significantly enhance overall performance. The research indicated that the SPL performance of this ceramic material in buzzers is close to that of similar products made from commercial PZT-4 materials (average 103.4 dB for KNNS-BNZ-1 mol% Al_2O_3 buzzers and average 103.7 dB for PZT-4 buzzers). Moreover, we found that this ceramic also exhibited superior atomization performance when used in atomizers. These findings could promote the development of lead-free piezoelectric ceramics.

2. Materials and methods

The conventional solid-state method was adopted to prepare the polycrystalline ceramics with composition $(0.96(K_{0.5}Na_{0.5})(Nb_{0.96}Sb_{0.04})O_3$–$0.04(Bi_{0.5}Na_{0.5})ZrO_3$–1 mol% Al_2O_3, abbreviated as KNNS-BNZ-1 mol% Al_2O_3). The raw materials used were K_2CO_3 (Sinopsin Group, 99.0%), Na_2CO_3 (Sinopsin Group, 99.8%), Nb_2O_5 (Sinopsin Group, 99.5%), Sb_2O_3 (Sinopsin Group, 99.99%), ZrO_2 (Sinopsin Group, 99.99%), Bi_2O_3 (Sinopsin Group, 99.999%) and Al_2O_3 (Sinopsin Group, 99.99%). All raw materials were weighed according to the stoichiometric ratio, after a dehydration treatment of K_2CO_3 and Na_2CO_3. Then, the raw materials were ground with ethanol as the mixing medium for 16 h in the ball mill tank. The dried and sifted mixture was put into a furnace at 850 °C for 6 h. After that, the mixture was dried and sifted after a second ball milling. Then, the refined powder was mixed with 3% PVA binder and pressed into a sheet at 8 MPa, which was discharged at 660 °C. The sheet was placed into a high-temperature furnace with the two-step sintering method. The sheet was heated up to 1180 °C at a rate of 10 °C/min and held for 5 min. After 3–5 min, the temperature dropped to 1060–1080 °C and was held for 15 h. The sintered ceramic sheet was polarized at room temperature with a polarization voltage of 3 kV/mm after being covered with silver electrodes on both sides, and the electrical properties were tested after polarization for 24 h. The commercial PZT-4 piezoelectric ceramics came from Audiowell Electronics (Guangdong) Co., Ltd.

A quasi-static piezo-d_{33} meter (ZJ-6A9, Chinese Academy of Sciences, China) was used to test the d_{33} value. The crystal and phase structure were analyzed using an x-ray diffractometer (XRD, X'pert pro) test. The surface morphology of the ceramics was tested by scanning electron microscope (SEM, Hitachi, S-4800). A transmission electron microscope (TEM, FEI Tecnai G2 F30) was used to observe the domain-wall structure. An impedance analyzer (4294A) was used to test the k_p and resonant frequency F_s. The dielectric constant of the ceramics was tested by a dielectric temperature spectrum test system (TZDM), with a testing frequency of 1 kHz and a heating rate of 2 °C/min. A ferroelectric tester (TZFE-300) was used to measure the P–E loop, with a testing frequency of 1 Hz and an electric field of 30 kV/cm. A strain tester (TZFE-300-s) was used to measure the S_{uni}–E loop, with a testing frequency of 50 Hz and a testing electric field of 30 kV/cm. The SPL test was conducted in an anechoic room, with a distance of 0.3 m between the sound source and the testing instrument. Testing of atomization quality was carried out in the laboratory and the atomization sheet was driven by a PCB circuit board.

3. Results and discussion

As the desire for an eco-friendly society continues to grow, lead-based ceramics will gradually be phased out from the realm of piezoelectric ceramics. For piezoelectric buzzers, the primary question is to determine which type of piezoelectric ceramic is suitable for their manufacture. To address this, we employed finite element analysis software to build a buzzer-air field model based on PZT-5A ceramic material, as shown in Figs. 1(a) and 1(b). We then simulated the physical parameters, including the piezoelectric constant, inverse piezoelectric constant and relative dielectric constant, as depicted in Figs. 1(c) and 1(d). The simulation results indicate that the inverse piezoelectric constant d_{33}^* of the piezoelectric material has the most significant impact on the SPL of the buzzer. The larger the inverse piezoelectric constant was, the better SPL performance the buzzer had. This provided clear guidance for designing and developing lead-free piezoelectric ceramic materials.

We studied the differences in dielectric, ferroelectric and strain between the KNNS-BNZ-1 mol% Al_2O_3 ceramics and the commercial PZT-4 ceramics, and the results are shown in Fig. 2. The temperature-dependent dielectric constants were separated and displayed in Figs. 2(a) and 2(b). It was obvious to see that the dielectric constant of the commercial PZT-4 ceramics was much higher than that of the KNN-based ceramic. In addition, there were two dielectric peaks in the

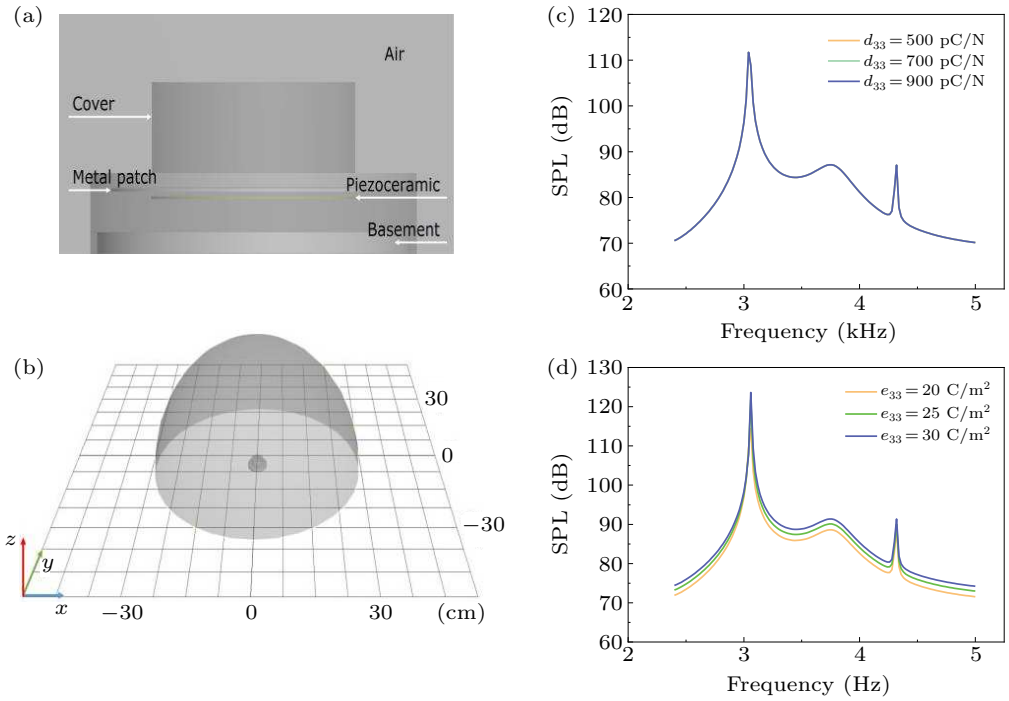


Fig. 1. Finite element analysis of piezoelectric buzzers. (a) Model construction, (b) buzzer–air field model, (c) the variation of sound pressure level with piezoelectric constant, (d) the variation of sound pressure level with inverse piezoelectric constant.

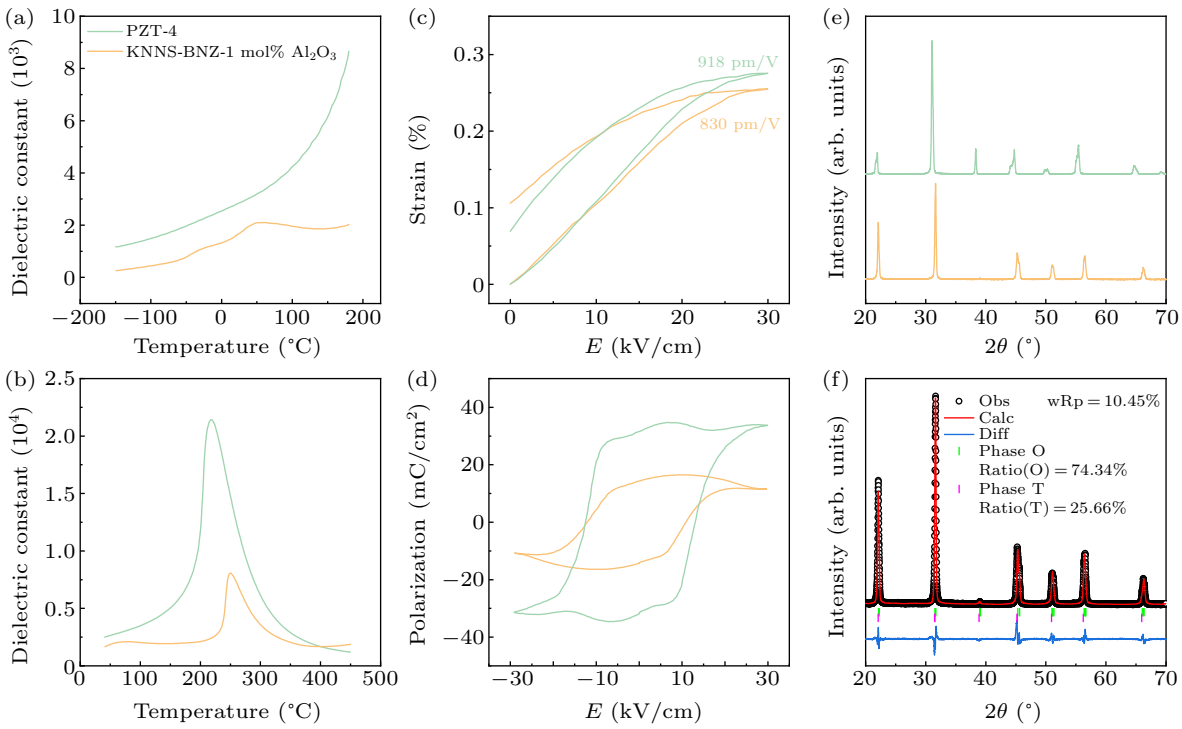


Fig. 2. Dielectric temperature spectrum in the (a) low-temperature region and (b) high-temperature region; (c) room-temperature P–E loop comparison and (d) room-temperature S_{uni}–E loop comparison between KNNS-BNZ-1 mol% Al_2O_3 and PZT-4 ceramics, (e) the XRD pattern of the KNNS-BNZ-1 mol% Al_2O_3 and PZT-4 ceramics; (f) the Rietveld refinement of the KNNS-BNZ-1 mol% Al_2O_3 ceramics.

KNNS-BNZ-1 mol% Al_2O_3 ceramics, which correspond to the (R–O) phase transition (the left peak, about −28 °C) and the (O–T) phase transition (the right peak, about 52 °C), respectively. Furthermore, the Curie temperatures of the KNNS-BNZ-1 mol% Al_2O_3 ceramics and the PZT-4 ceramics were 250 °C and 218 °C, respectively. The dielectric constant reflected the polarization ability of the medium, and the larger the dielectric constant value, the stronger the polarization ability was;[24] this was consistent with the P–E loops, which are displayed in Fig. 2(c). The coercive fields of the two kinds of ceramics were close. Whereas the maximum polarization (P_{max}) of the commercial PZT-4 ceramic was almost

two times that of the KNNS-BNZ-1 mol% Al_2O_3 ceramics (34.64 μC/cm^2 for the PZT-4 ceramic and 16.36 μC/cm^2 for the KNNS-BNZ-1 mol% Al_2O_3 ceramic), which was in agreement with the former analysis. Figure 2(d) shows the uni-polar strains of the two ceramics under an electric field of 30 kV/cm; both of the two ceramics exhibit similar d^*_{33} (830 pm/N for the KNNS-BNZ-1 mol% Al_2O_3 ceramics and 917 pm/N for the PZT-4 ceramics), indicating that the KNNS-BNZ-1 mol% Al_2O_3 ceramics possessed good application performance compared with the commercial PZT-4 ceramics.

Figures 2(e) and 2(f) showed the crystal structure data of the un-polarized KNNS-BNZ-1 mol% Al_2O_3 ceramics and commercial PZT-based ceramics collected by the XRD machine, both showing typical characteristics of the perovskite structure. Typically, the phase structure of KNNS-BNZ-1 mol% Al_2O_3 ceramics was initially judged by the relative intensities of the (002) and (200) Bragg peaks (about 45°).[25] In order to clarify the occupation fraction and the cell parameters, Rietveld refinement of the KNNS-BNZ-1 mol% Al_2O_3 sample was carried out by GSAS software and the refinement map, phase fraction and corresponding cell parameters were obtained, which are displayed in Fig. 2(f) and Table 1. The fractions of orthorhombic and tetragonal phases were 74.34% and 25.66%, respectively. Moreover, the R_{wp} was 10.45%, which indicates that the consequence of refinement was reliable. The O–T two-phase coexistence made KNNS-BNZ-1 mol% Al_2O_3 ceramics have excellent properties ($d_{33} = 480$ pC/N, $k_p = 0.62$).

Besides the lattice contribution, the domain-wall morphology also had a significant impact on the improvement of piezoelectric performance of ferroelectric materials. Figure 3 shows the morphology, domain-wall and grain size distribution of the KNNS-BNZ-1 mol% Al_2O_3 ceramics and the PZT-4 ceramics. It is clear to see two kinds of domains in Fig. 3(a). The white circle represents the striped domain and the yellow circle represents the polar nanoregions (PNRs), which could promote the domain boundary switching due to the flattened free energy,[26–31] indicating that the KNNS-BNZ-1 mol% Al_2O_3 ceramics had good piezoelectric properties. Otherwise, there was no significant domain structure in the PZT-4 ceramics in Fig. 3(b), but we surprisingly found that there were many tiny domains at the nanoscale, which indicates the high piezoelectric constant (about 500 pC/N). Both kinds of ceramics had relatively dense micro-morphology, as depicted in Figs. 3(c) and 3(d). According to the grain size distribution results, the KNNS-BNZ-1 mol% Al_2O_3 ceramic had a smaller average grain size (2.41 μm for the KNNS-BNZ-1 mol% Al_2O_3 ceramic and 3.58 μm for the PZT-4 ceramic), but the uniformity of the grain size was inferior to that of PZT-4 ceramics. This may be due to the difference in the synthesis process of the two kinds of ceramics. The PZT-4 ceramics were synthesized by the tape casting method, which made the ceramics have more uniform grain size and finally led to better consistency in batch production of the ceramics.

Table 1. Cell parameters of KNNS-BNZ-1mol% Al_2O_3 ceramics.

KNNS-BNZ-1 mol% Al_2O_3		
Phase	O	T
Ratio (%)	74.34	25.66
a (Å)	3.9606	3.9944
b (Å)	5.6276	3.9944
c (Å)	5.6430	3.9951

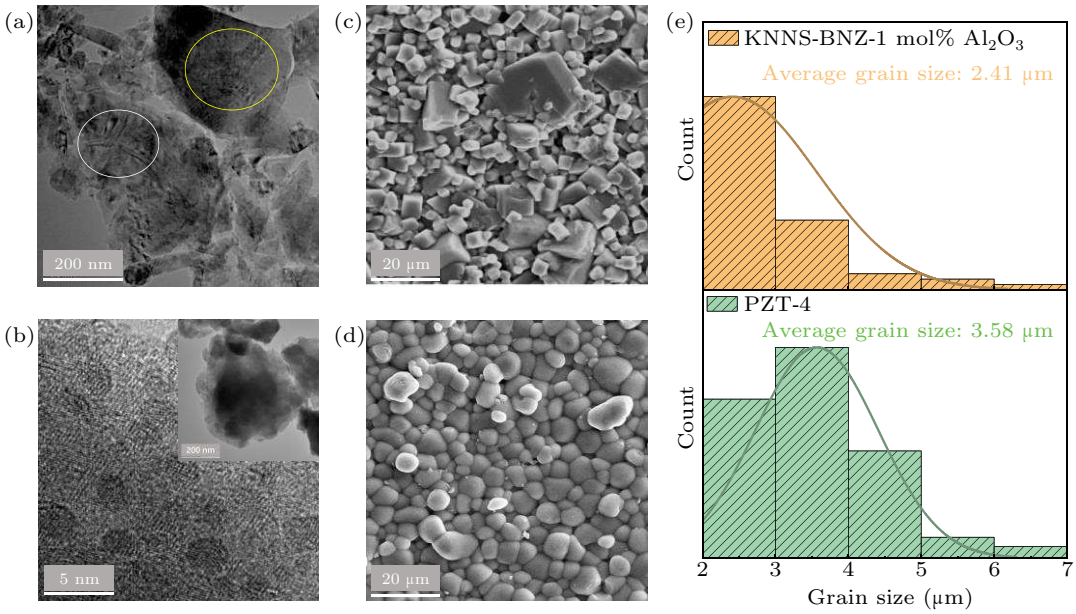


Fig. 3. Transmission electric microscope (TEM) bright-field images of (a) KNNS-BNZ-1mol% Al_2O_3 ceramics and (b) PZT-4 ceramics. Scanning electron microscope (SEM) images of (c) KNNS-BNZ-1mol% Al_2O_3 and (d) PZT-4 ceramics, (e) grain size distribution and variation of average grain size of the two kinds of ceramics.

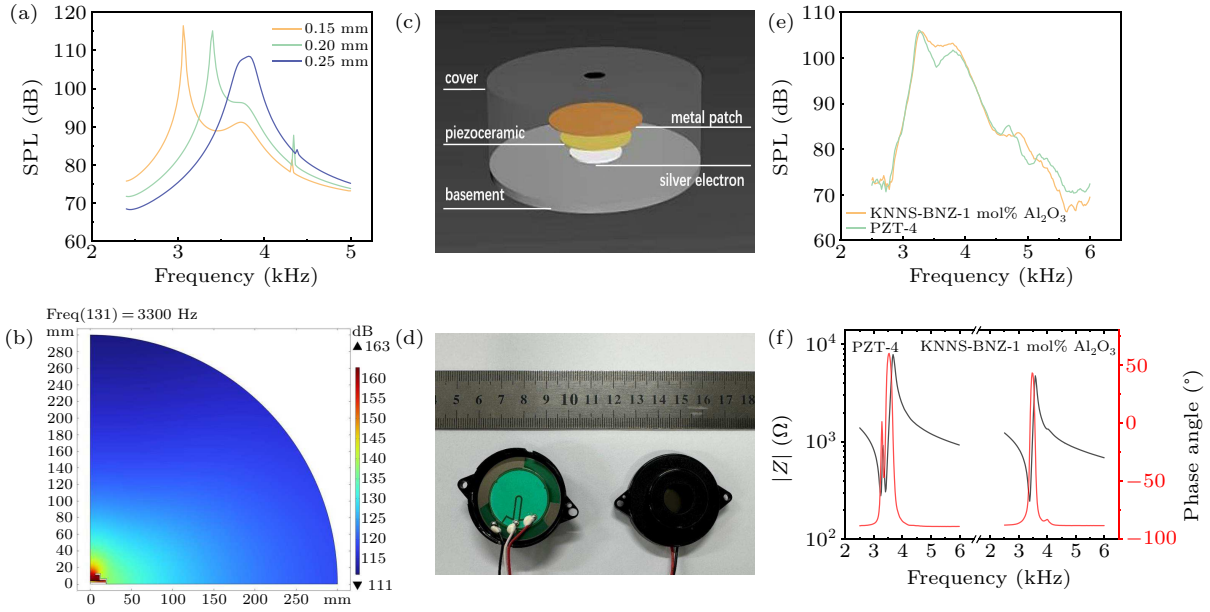


Fig. 4. Finite element analysis, fabrication and performance of piezoelectric buzzers. (a) The sound pressure level performance of KNN-based piezoelectric buzzers with different ceramic thicknesses, (b) overall sound pressure level performance of the 0.20 mm buzzer, (c) structure schematic of buzzer; (d) demo of the KNN-based buzzer, (e) the SPL curves of two kinds of buzzers under a driving voltage of 3.5 Vrms, (f) frequency dependence of $|Z|$ and θ for two kinds of buzzers.

Table 2. Main parameters comparison between five KNN-based and PZT-4 buzzers.

	SPL (dB)		Peak value of SPL (dB)		Resonant frequency (kHz)		R_1 (Ω)		Capacitance (pF)	
	KNN	PZT-4	KNN	PZT-4	KNN	PZT-4	KNN	PZT-4	KNN	PZT-4
	102.8	103.7	108.9	103.7	3.48	3.35	298	229	46940	35120
	104.4	103.5	106.8	104.5	3.39	3.34	272	216	45900	34700
	102.8	103.5	105.8	106.0	3.45	3.32	311	230	44420	34830
	103.5	103.8	105.8	103.0	3.38	3.38	302	330	45240	35250
	103.3	103.8	107.5	105.4	3.40	3.31	253	235	42770	36100
Average	103.4	103.7	107.5	105.4	3.40	3.31	287	248	45054	35200

The development of piezoelectric buzzers has a history of over a hundred years and its manufacturing process has nearly reached perfection. To reduce production costs, we considered designing KNN-based ceramic buzzers based on the original PZT ceramic buzzers. Firstly, we conducted finite element analysis using COMSOL software to determine the optimal ceramic thickness and all of the factors of the KNNS-BNZ-1mol% Al_2O_3 ceramic came from the Ref. [32]. As shown in Fig. 4(a), we could see that when the ceramic thickness was 0.20 mm, the buzzer exhibited the best SPL performance. Figure 4(b) shows the overall SPL performance of the 0.20 mm buzzer. Based on the above results, a KNNS-BNZ-1 mol% Al_2O_3 ceramic buzzer was fabricated. First, the sintered KNNS-BNZ-1 mol% Al_2O_3 ceramic was cut into circular thin sheets with a radius of 1.25 cm and a thickness of 0.20 mm. The sheets were then processed with silver coating and polarization to create piezoelectric diaphragms. Subsequently, the diaphragms were assembled with other components to form the piezoelectric buzzer, followed by performance testing.

Figures 4(c) and 4(d) show the simple structure of the buzzer and a demo picture of the buzzer made using KNNS-BNZ-1 mol% Al_2O_3 piezoelectric ceramics. The piezoelectric buzzers were fabricated to compare the SPL under a driving voltage of 3.5 Vrms, as shown in Fig. 4(e). Throughout the entire sweep frequency test range, both buzzers maintained an SPL above 70 dB, which exhibited practical application potential. Overall, the KNNS-BNZ-1 mol% Al_2O_3 buzzer showed a comparable SPL to the PZT-4 buzzer and even outperformed it in the peak range of 3–4 kHz. Then, we tested the impedance and phase angle dependence on the frequency of the two kinds of buzzers, as depicted in Fig. 4(f), and found that the two buzzers had approximately similar impedance, phase angle and resonant frequency. Then, we tested the resonant frequency, the SPL performance, the resistance and the capacitance of the two kinds of buzzers, as displayed in Table 2. The SPL performance of the two kinds of buzzers was very close and excellent, which shows the possibility of replacing the PZT-4 ceramics in buzzer applications. Furthermore, the buzzers made by KNNS-BNZ-1 mol% Al_2O_3 ceramics had a better peak value of SPL than the buzzers made using PZT-4 ceramics.

Furthermore, we also explored the possibility of using the KNNS-BNZ-1 mol% Al_2O_3 piezoelectric material for an atomizer. We adopted radial vibration mode and designed a microporous atomizer, as depicted in Fig. 5. The resistance R_1 and capacitance are the main parameters of the piezoelectric ceramics in the buzzer and atomizer applications. The resistance R_1 is related to the heat generation of piezoelectric ceramics and low resistance is beneficial for improving the service life of piezoelectric ceramics.[33] In addition, the capacitance reflects the performance of the piezoelectric ceramics. Several main factors and the test results of the two kinds of atomizers are listed in Table 3. In the test, we adopted an intermittent atomization test method, which involved atomizing for one minute, pausing for two minutes and finally accumulating atomization for one hour. We found that the R_1 and capacitance of the KNNS-BNZ-1 mol% Al_2O_3 atomizer (around 95 Ω and 5500 pF, respectively), were better than those of the PZT-4 atomizer (around 180 Ω and 2700 pF, respectively), which led to a better atomization quality. This may be important progress in the exploration of lead-free piezoelectric ceramic applications.

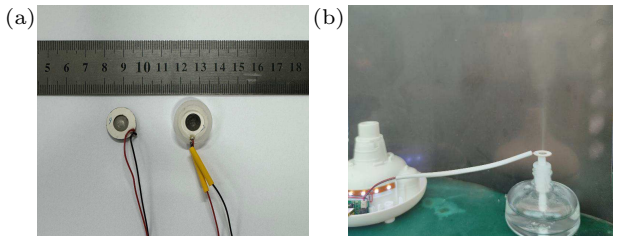


Fig. 5. (a) Actual atomization components manufactured using the 0.96KNNS-0.04BNZ-1 mol% Al_2O_3 ceramics; (b) the atomization quality test.

Table 3. Main factors comparison between the KNNS-BNZ-1 mol% Al_2O_3 and PZT-4 atomizers.

	Resonant frequency (kHz)	R_1 (Ω)	Capacitance (pF)	Atomization quality (mL/min)	Power (W)
KNN1*	139.96	94	5522		
KNN2	140.35	98	5404	Average: 0.8	3.0
KNN3	139.29	94	5613		
PZT4	108	< 180	2700	0.64	1.2

*KNN1, KNN2, and KNN3 are the designations of the fabricated atomizers, which were prepared using the same material.

4. Conclusion

We have developed a buzzer based on lead-free piezoelectric ceramic materials of 0.96KNNS-0.04BNZ-1 mol% Al_2O_3 prepared by the solid-state reaction method. The coexistence of the O–T phase and the stripe domain promoted its excellent piezoelectric performance. More importantly, the outstanding SPL performance (average 103.4 dB) of the buzzer and good atomization quality of the atomizer (average 0.8 mL/min) revealed the enormous application potential for replacing the PZT-4 piezoelectric ceramics in special fields.

Acknowledgments

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