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Research Article

Fe-doped BNKBT lead-free piezoceramics with enhanced electromechanical performance for 10 MHz ultrasonic imaging

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Abstract: Ultrasonic transducers have long relied on lead-based piezoelectric materials. However, the potential harm of lead to the environment and human health has spurred an urgent demand for environmentally friendly lead-free alternative materials. Bismuth sodium titanate (BNT) is regarded as a promising piezoelectric material for lead-free transducers due to its low dielectric constant and acoustic impedance. To address the problem that the previously developed 0.84BNT-0.133BKT-0.027BT composition still struggles to meet the requirements of high-quality biological tissue imaging, this study adopted an acceptor doping strategy and prepared a B-site Fe^{3+} -doped lead-free piezoceramic system. The results show that appropriate Fe^{3+} doping produces a favorable balance between $R3c/P4bm$ phase-structure modification and defect-related pinning, thereby enhancing the electromechanical response. At $x = 0.01$, the piezoelectric coefficient reaches the maximum value of 180 pC/N, while k_t reaches 0.42, indicating enhanced thickness-mode electromechanical coupling. The planar 10 MHz ultrasonic transducer fabricated with this composition exhibits high sensitivity, with an insertion loss $|IL|$ of 16 dB. After introducing an acoustic focusing lens, the focused transducer achieves axial and lateral resolutions of 182 and 264 μm , respectively. In addition, clear imaging of the internal structure of a porcine eyeball and the surface of a commemorative coin was successfully achieved. These results confirm that the B-site Fe^{3+} -doped modified BNT-BKT-BT ternary lead-free ceramic has great application potential in high-performance environmentally friendly medical ultrasonic transducers.

Keywords: Bismuth sodium titanate (BNT); lead-free piezoceramic; Fe^{3+} -doped; piezoelectric properties; ultrasonic transducer.

1 Introduction

Ultrasound imaging technology plays a vital role in current medical diagnosis due to its real-time and non-invasive characteristics, and has been widely used in medical examination of human tissues such as eyes, heart and liver [1-4]. In ultrasound imaging, the performance of ultrasonic transducer

determines the image quality, and as the key functional component responsible for energy conversion in ultrasonic transducers, the performance of piezoelectric materials has an important impact on the performance of transducers [5,6]. At present, lead-based piezoelectric materials represented by lead zirconate titanate (PZT) ceramics and related lead-based perovskites dominate the market due to their excellent piezoelectric properties. However, these lead-based piezoelectric materials contain a large amount of toxic lead element, which poses potential threats to the ecological environment and human health [7,8]. With the increasing public awareness of environmental protection, various countries have successively introduced relevant regulations restricting the use of lead in electronic products. Therefore, developing new lead-free piezoelectric materials with both excellent piezoelectric properties and environmental friendliness, and promoting the lead-free transformation of ultrasonic transducers, have important scientific significance and urgent market demand for the sustainable development of ultrasonic transducers. Among many lead-free piezoelectric material systems, perovskite-type lead-free piezoceramics have attracted much attention due to their high piezoelectric activity and tunable phase structure. Among them, $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ (BNT)-based piezoceramics are considered one of the strong candidates to replace lead-based materials for ultrasonic transducers because of their low dielectric constant and low acoustic impedance [9]. However, pure BNT ceramics have high coercive fields and low piezoelectric performance, which limit the energy conversion efficiency, and further restrict application for ultrasonic transducers [10].

In previous research, we selected BaTiO_3 (BT) and $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ (BKT) to form a multi-component solid solution BNT-BKT-BT with BNT. By adjusting the proportion of BNT, we constructed a morphotropic phase boundary (MPB) between the relaxor phase and the ferroelectric phase, which significantly improved the ferroelectric and piezoelectric properties of BNT ceramics [11]. On this basis, we selected 0.84BNT-0.133BKT-0.027BT ceramics with the critical MPB composition and successfully prepared a lead-free piezoelectric ultrasonic transducer with a center frequency (f_c) of 10 MHz. Although this transducer has good frequency band response and high sensitivity, its performance

still has a certain gap from meeting the practical application requirements of ultrasonic imaging for biological tissues. Many studies have found that the composition at MPB can still adjust the Gibbs free energy curve of its system through several strategies, so as to further optimize the ferroelectric and piezoelectric properties of ceramics. These strategies include ion doping modification [12-15], AC polarization treatment [16], grain textured growth [17,18], and electric/thermal-field regulation [19]. When traditional PZT piezoceramics with perovskite structure are doped with B-site acceptors, negatively charged defects will be generated in the crystal lattice, accompanied by the generation of oxygen vacancies for charge compensation. Affected by this, the material presents typical hard piezoelectric properties: improved temperature stability, reduced dielectric loss, decreased piezoelectric constant, and increased coercive field [20]. When acceptor doping is performed on BNT-based piezoceramics, the hard piezoelectric effect exhibited by the ceramics is slightly different. For example, in 1 at% Fe^{3+} -doped BNT and 0.8BNT-0.2BKT, although defect dipoles that cause piezoelectric effect hardening can be detected, the coercive field only increases slightly [21,22]; when Mn^{2+} replaces Ti^{4+} in BNT ceramics, it will increase the lattice distortion degree of the rhombohedral ferroelectric phase in BNT ceramics. The ferroelectricity of the ceramics is significantly improved, and the dielectric loss of the ceramics is significantly reduced, but the depolarization temperature of the ceramics decreases slightly [23]; For 0.9BKT-0.06BT-0.04BLT ceramics doped with 1 wt% Cu^{2+} , not only is the sintering temperature required reduced, but its piezoelectric properties and electromechanical coupling ability are also improved [24]. These differences mainly originate from the unique “relaxor phase-ferroelectric phase” polymorphic evolution in BNT-based piezoceramics. The volume fraction of materials that can be stabilized by internal defect dipoles is lower than that of PZT piezoceramics, resulting in a weaker hard piezoelectric effect [25].

In the periodic table of elements, iron and titanium both belong to the fourth-period transition group with close atomic numbers. There is only one positive charge difference between Fe^{3+} and Ti^{4+} , and Fe^{3+} has hexacoordination ability, tending to replace Ti^{4+} at the B-site [26]. Therefore, this work takes

the 0.84BNT-0.133BKT-0.027BT ceramic at the critical MPB composition obtained in our previous work as the matrix, designs and prepares a B-site Fe^{3+} acceptor doped substituted piezoceramic composition system of $(\text{Bi}_{0.487}\text{Na}_{0.42}\text{K}_{0.067}\text{Ba}_{0.027})(\text{Ti}_{1-x}\text{Fe}_x)\text{O}_3$, and systematically investigates the influence of Fe doping content on the phase composition, microscopic morphology, dielectric properties, domain structure, ferroelectric, and piezoelectric properties of the ceramics. The research and design scheme is shown in Fig. S1. Ultrasonic transducers operating near 10 MHz provide a suitable balance between spatial resolution and penetration depth, making them attractive for ophthalmic imaging, superficial-tissue examination, and imaging of small anatomical structures. They are also applicable to high-resolution nondestructive inspection of small components and surface features. Therefore, based on the ceramic with optimal performance in the research system, an ultrasonic imaging transducer with a center frequency of 10 MHz was designed and fabricated, and relevant performance tests and imaging application research were carried out.

2 Experimental

2.1 Preparation of lead-free piezoceramics and transducers

Lead-free piezoceramics $(\text{Bi}_{0.487}\text{Na}_{0.42}\text{K}_{0.067}\text{Ba}_{0.027})(\text{Ti}_{1-x}\text{Fe}_x)\text{O}_3$ [abbreviated as BNKBT-Fe(x) with $x = 0, 0.005, 0.01, 0.015, 0.02, 0.03$] were prepared by the conventional solid-state method. Raw materials Bi_2O_3 (99.9%), Na_2CO_3 (99.99%), K_2CO_3 (99.98%), BaCO_3 (99.95%), TiO_2 (99.9%), and Fe_2O_3 (99.95%) were weighed according to the designed formula, then ball-milled for 12 h in a planetary ball mill with anhydrous ethanol and zirconia grinding balls as milling media. Subsequently, the milled slurry was placed in a constant temperature drying oven and dried at 80 °C. The dried powder was then pressed into a block and placed in a muffle furnace to calcine at 850 °C / 2 h to complete the pre-sintering reaction. After that, the pre-sintered block was crushed and subjected to a second ball milling following the same grinding procedure. The dried powder was pressed into white disc-shaped green compacts via cold isostatic pressing, and finally sintered at 1130 °C / 2 h. Silver electrodes were then coated on both sides of the BNKBT-Fe(x) ceramics and heat-treated for dielectric,

ferroelectric, and piezoelectric measurements. After silver electrode coating, the ceramics were immersed in insulating silicone oil for polarization treatment. The complete poling conditions: poling temperature = 120 °C, DC poling electric field = 5 kV/mm, poling duration = 15 min.

A target center frequency (f_c) of 10 MHz was selected to achieve a practical balance between imaging resolution and penetration depth for ophthalmic and superficial-tissue imaging, as well as small-scale nondestructive inspection. Based on the BNKBT-Fe(0.01) piezoceramic, an ultrasonic transducer with a center frequency (f_c) of 10 MHz was further designed and fabricated. First, the structural parameters of the ultrasonic transducer were designed and optimized using the simulation software PiezoCAD. The thickness of the BNKBT-Fe(0.01) piezoceramic was ground to 230 μm with a tolerance of 1 μm . Gold electrodes were fabricated on the upper and lower surfaces of the thinned ceramic using a DC sputtering coater. An acoustic matching layer and a backing layer were then fabricated on the upper and lower surfaces, respectively. After molding, the blank was processed into a cylinder with a diameter of 4.65 mm and a height of 2 mm on a lathe (Black Box CNC, CHN). The processed cylinder was then encapsulated in a brass housing. Finally, a gold electrode was fabricated on the entire front face of the transducer through the same sputtering system. To optimize the imaging performance of the transducer, an acoustic lens with a radius of curvature of 4.5 mm was mounted on the front end of the transducer to focus the acoustic beam of the transducer.

2.2 Characterization

The phase structure of the ceramics was characterized by X-ray diffractometer (XRD, D8 ADVANCE, Bruker, Germany). The surface morphologies were observed using a scanning electron microscope (SEM, SU8600, Hitachi, Japan). The ferroelectric and electrostrain properties were measured at 1 Hz using a ferroelectric analyzer (TF Analyzer 3000, aixACCT, Germany). The piezoelectric coefficient

(d_{33}) was tested by a quasi-static d_{33} meter (J-6BN, Institute of Acoustics, Chinese Academy of Sciences, Beijing, China). The domain structures were characterized by piezoresponse force microscopy (PFM, Cypher ES, Oxford Instruments Asylum Research, USA). The topography, amplitude, and phase signals were acquired simultaneously with 256×256 pixels. The scan area was $5 \times 5 \mu\text{m}^2$. The scan rate was 1-2 Hz, and a dual-frequency AC excitation of 1 V was applied. X-ray photoelectron spectroscopy (XPS, VersaProbe 4, ULVAC-PHI, Japan) was used to analyze the surface chemical states, including lattice- and defect-related oxygen of the ceramics. The dielectric properties were measured by a precision LCR meter (TH2838A, Shanghai, China). The resonance and antiresonance frequencies were measured using an impedance analyzer (WK6500B, Wayne Kerr Electronics, UK). The thickness-mode electromechanical coupling factor (k_t) and planar radial-mode electromechanical coupling factor (k_p) were calculated using the following resonance-antiresonance equations:

$$c = 2 d f_r^t \quad (1)$$

$$k_t = \sqrt{\frac{\pi f_r^t}{2 f_a^t} \tan\left(\frac{\pi f_a^t - f_r^t}{f_a^t}\right)} \quad (2)$$

$$k_p = \frac{1}{\sqrt{0.398 \frac{f_r^p}{f_a^p - f_r^p} + 0.579}} \quad (3)$$

Where (c) is the longitudinal acoustic velocity along the thickness direction; (d) is the thickness of the ceramic; (f_r^t) and (f_a^t) are the resonance and antiresonance frequencies of the thickness-extensional vibration mode, respectively; and (f_r^p) and (f_a^p) are those of the planar radial vibration mode, respectively. All BNKBT-Fe(x) ceramics had an approximately identical diameter of 9 mm and thickness of 0.5 mm, and were characterized using the same measurement procedure. Therefore, (k_t/k_p) was used to compare the relative prominence of thickness-mode coupling over radial-mode coupling within the present composition series.

For the ultrasonic transducer characterization, the impedance and phase angle of the transducer were measured with an impedance analyzer (WK6500B, Wayne Kerr Electronics, UK) to evaluate 50 Ω impedance matching at the center frequency (f_c). The phase-curve peak reflects the phase lag between voltage and current, indicating the actual electric power obtained by the transducer. Pulse-echo measurements were conducted in deionized water, using a quartz plate as the reflector. The reflector was positioned at a distance providing the maximum echo amplitude. An Olympus 5073PR pulser/receiver (Olympus, USA) with a 50 Ω output impedance, a pulse-repetition frequency of 200 Hz, and a pulse energy of 10 μ J was used to excite the transducer and receive the echo signal. The waveform was recorded using a DSOX3024A oscilloscope (Keysight, USA). The insertion loss (IL) was measured using a DG1022 function generator (RIGOL, China) and a DSOX3024A oscilloscope (Keysight, USA). A 20-cycle sinusoidal burst at the center frequency of the transducer was used as the excitation signal. The transducer position was adjusted until the maximum echo amplitude was obtained. The oscilloscope input impedance was first set to 1 M Ω to measure the received echo voltage, V_1 . The transducer was subsequently disconnected, and the oscilloscope input impedance was set to 50 Ω to measure the reference excitation voltage, V_2 . The insertion loss was calculated as:

$$IL = 20 \cdot \log \frac{V_1}{V_2} + \alpha_w \cdot 2d_w \cdot f^2 + 1.9 \quad (4)$$

$$d_w = \frac{c_w \cdot t}{2} \quad (5)$$

where V_1 , V_2 are echo and input signal amplitudes, f is excitation frequency; α_w is water attenuation coefficient, $2.2 \times 10^{-4} \text{ dB} \cdot (\text{mm} \cdot \text{MHz}^2)^{-1}$, d_w is transducer-to-reflector distance, 1.9 dB is quartz-plate reflection loss, c_w is sound velocity in water, and t is echo arrival time. Resolution measurements were performed using a DPR 500 pulser/receiver (JSR Ultrasonics, USA) with a 50 Ω output impedance, a pulse-repetition frequency of 200 Hz, and a pulse energy of 12.4 μ J. Three tungsten wires with a diameter of 100 μ m were fixed in the water tank, and the transducer was mounted on a motorized

three-axis scanning stage. For B-mode imaging, the scanning step was 4 $\mu\text{m}/\text{step}$ along the X direction.

For C-mode imaging, the scanning steps were 4 and 100 $\mu\text{m}/\text{step}$ along the X and Y directions.

3 Results and Discussion

3.1 Analysis of phase structure and microscopic morphology

Fig. S2(a) shows the XRD patterns of BNKBT-Fe(x) ceramics. All ceramics exhibit pure perovskite diffraction peaks without any impurity phases, demonstrating that Fe^{3+} doping does not break the complete perovskite lattice during sintering. From the amplified (111) and (200) peak positions in Fig. S2(b) and (c), it can be seen that the peak positions shift to the left. This shift corresponds to an increase in the interplanar spacing and is consistent with the substitution of high-spin Fe^{3+} for Ti^{4+} at the octahedral B-site of the perovskite lattice [27,28]. To further clarify the phase evolution, Rietveld refinement was performed using a two-phase model consisting of rhombohedral ($R3c$) and tetragonal ($P4bm$) phases, as shown in Fig. S3 and Table S1. The calculated profiles agree well with the experimental patterns, with (R_{wp}) values $< 10\%$ and GOF values < 2 for all ceramics, indicating that the two-phase model reasonably describes the experimental XRD patterns. Overall, increasing Fe dopant drives the structural transformation from $R3c$ -dominated to $P4bm$ -dominated state; a slight rise in $R3c$ phase is observed at $x = 0.02$, and the ceramic becomes strongly $P4bm$ -dominated at $x = 0.03$. Fig. S4 shows SEM images of the grain morphology of BNKBT-Fe(x) ceramics after thermal corrosion treatment. Growth striations originating from crystal growth can be observed on grain surfaces. Quantitative grain-size analysis was performed, and the corresponding grain-size distribution histograms and average grain size (AGS) for each composition are also presented in Fig. S4. With increasing x , the AGS increases first and then decreases, and the grain size gradually becomes more uniform, suggesting that Fe doping promotes the sintering behavior of the ceramics [27].

3.2 Dielectric, domain, ferroelectric, and piezoelectric properties

The temperature and frequency-dependence of the dielectric constant (ϵ_r) and loss tangent ($\tan \delta$) of poled BNKBT-Fe(x) ceramics are shown in Fig. 1. All ceramics exhibit three dielectric anomalies: (1)

A ferroelectric-to-relaxor transition exists in the low-temperature ($45 \sim 105^\circ\text{C}$), corresponding to the depolarization temperature (T_d); (2) There is obvious dielectric relaxation in the medium temperature ($T_d \sim 200^\circ\text{C}$), and significant frequency dispersion occurs in this temperature range; this temperature is defined as T_s ; (3) A structure phase transition occurs in the high-temperature ($300 \sim 350^\circ\text{C}$), where the ferroelectric disappears, and the ε_r reaches the maximum value at this time; this temperature is defined as T_m . Furthermore, the T_d values marked in Fig. 1 show a gradually increasing trend with increasing Fe^{3+} content. This phenomenon can be attributed to the acceptor-type substitution of Ti^{4+} by Fe^{3+} , and the oxygen vacancies introduced into the lattice will increase the T_d [21,29,30].

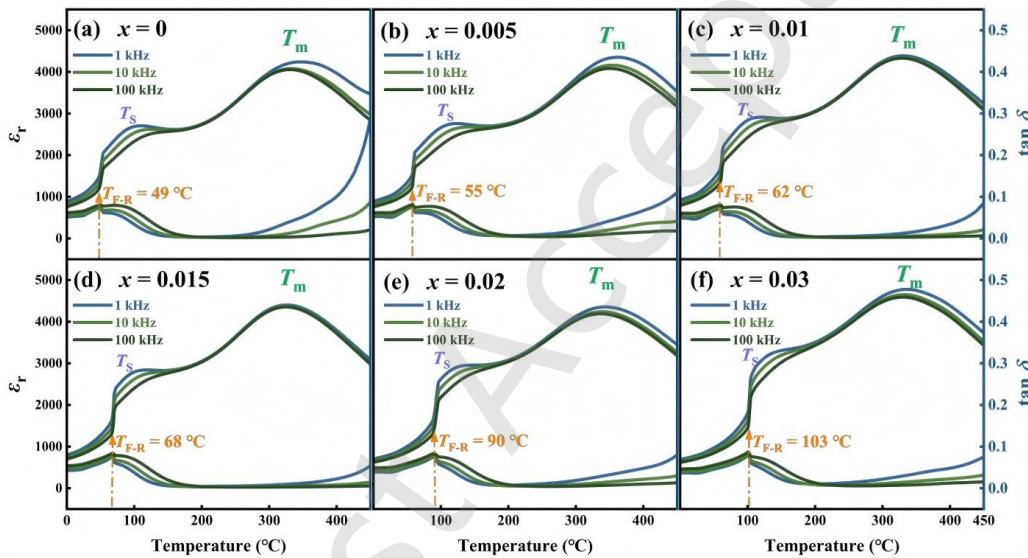


Fig. 1. Temperature-frequency dependence of ε_r and $\tan \delta$ of polarized ceramics.

The domain structure of unpoled BNKBT-Fe(x) ceramics was investigated by PFM, as shown in Fig. 2. For the undoped ceramic $x = 0$, the domain distribution is relatively discrete, and large-area non-polar disordered relaxor phase regions can be observed, as shown in Fig. 2(a₁-c₁). This feature may be related to the local compositional and structural heterogeneity characteristic of BNT-based ceramics [9,31]. The $x = 0.01$ ceramic exhibits larger and more clearly defined domains than the undoped, as shown in Fig. 2 (a₂-c₂), indicating that moderate Fe doping modifies the local domain configuration. At $x = 0.02$, the domain distribution becomes uniform, whereas the amplitude response is markedly

suppressed, suggesting a restricted local electromechanical response. Local polarization switching was further evaluated by applying a DC bias voltage to write a “square-loop” domain pattern, as shown in Fig. 2 (a₃-c₃). The $x = 0$ ceramic exhibits limited polarization switching ability, the boundaries of the inner and outer squares are relatively blurred, and some domains cannot maintain a stable switched state. This is mainly attributed to the presence of relaxor regions in the written area. These regions can transform into a ferroelectric phase under an external electric field, but revert to the weakly polar or non-polar state after removal of the field. For $x = 0.01$, the pattern shows a clearer boundary and uniform phase reversal. The clearer written pattern and more uniform phase reversal at ($x = 0.01$) indicate an enhanced local polarization-switching response [31,32]. When $x = 0.02$, the pattern is almost completely blurred, no continuous polarized region is formed at the periphery, and the internal phase contrast is low, indicating that the domains cannot switch by the voltage applied from the probe. The phase-voltage and amplitude-voltage hysteresis loops for $x = 0, 0.01$, and 0.02 , are shown in Fig. 2(a₄-c₄). The results show that appropriate Fe doping can significantly improve the phase switching ability and strain response. At $x = 0.01$, the phase difference generated by domain reversal reaches 171° , and the maximum amplitude reaches 387 pm, which are improved compared to $x = 0$. However, oxygen vacancies may be generated for charge compensation even at a low Fe content when Fe^{3+} substitutes for Ti^{4+} . With increasing Fe content, oxygen-vacancy-related defects may interact more strongly with domain walls and act as local energetic pinning centers, thereby increasing the energy barrier for domain-wall motion [22,33]. This interpretation is consistent with the inhibited phase switching and reduced strain response observed at $x = 0.02$.

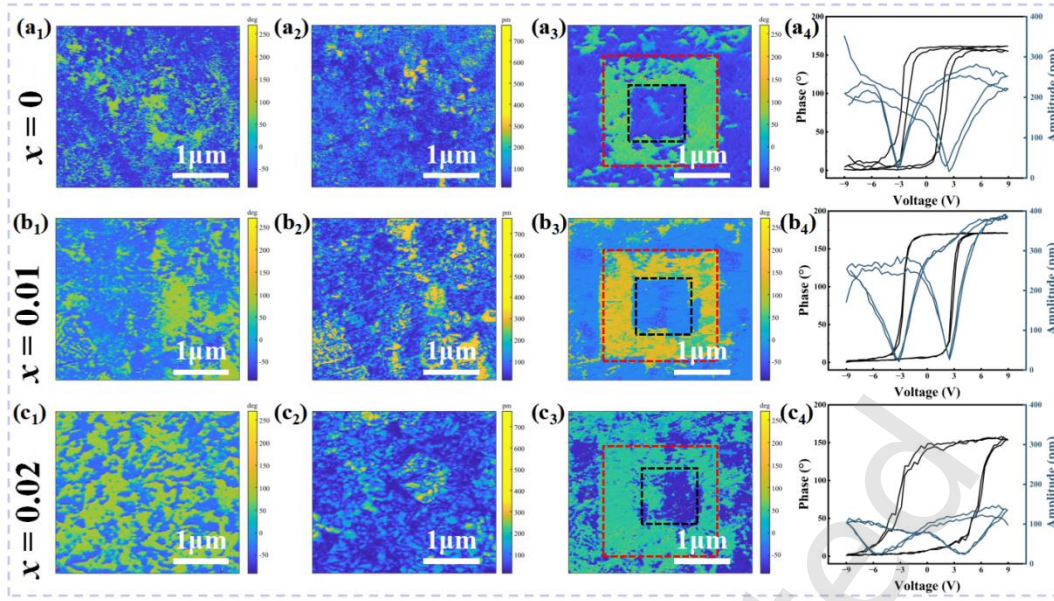


Fig. 2. PFM characterization of unpoled BNKBT-Fe(x) piezoceramics: (a₁-a₄) $x = 0$; (b₁-b₄) $x = 0.01$; (c₁-c₄) $x = 0.02$. Panels (a₁, b₁, and c₁), (a₂, b₂, and c₂), (a₃, b₃, and c₃), and (a₄, b₄, and c₄) show the out-of-plane PFM phase images, amplitude images, DC-bias-written domain patterns, and local phase-voltage (black) and amplitude-voltage (blue) responses, respectively. The red and black dashed boxes indicate the outer and inner written regions, respectively.

The bipolar polarization-electric field (P - E) loops and current density-electric field (J - E) curves of BNKBT-Fe(x) ceramics were measured at room temperature under 60 kV/cm and 1 Hz frequency, as shown in Fig. 3. When $x = 0$ and 0.005, the P - E loops show a relatively saturated state, whereas the J - E curves display diffuse and broadened peaks near the coercive field. This indicates that dipole polarization switching occurs over a broad electric-field range without abrupt changes, reflecting a polarization response influenced by relaxor behavior [9,31,34]. The maximum current density decreases from 4.08 mA/cm² ($x = 0.0$) to 3.28 mA/cm² ($x = 0.005$). This indicates that Fe doping modifies the field-induced polarization response. The $x = 0.01$ ceramic exhibits typical ferroelectric behavior, with a saturated P - E loop and a high polarization state. The remnant polarization (P_r) and maximum polarization ($P_{\max}$) reach 33 $\mu\text{C}/\text{cm}^2$, and 42 $\mu\text{C}/\text{cm}^2$, respectively. Meanwhile, the diffuse peaks in the J - E curve merge into a sharp current peak (J^{peak}) near the coercive field, indicating more synchronized domain switching under a critical electric field [35]. As the Fe content increases from x

$x = 0.01$ to 0.03 , the P - E loop remains saturated, P_r and $P_{\max}$ remain unchanged. However, the coercive field gradually increases, and the J - E curves show an obvious single-peak characteristic. This behavior may be associated with the increased concentration of oxygen-vacancy-related defects at higher Fe contents, which hinders domain-wall motion and increases the energy barrier for polarization switching [22,36].

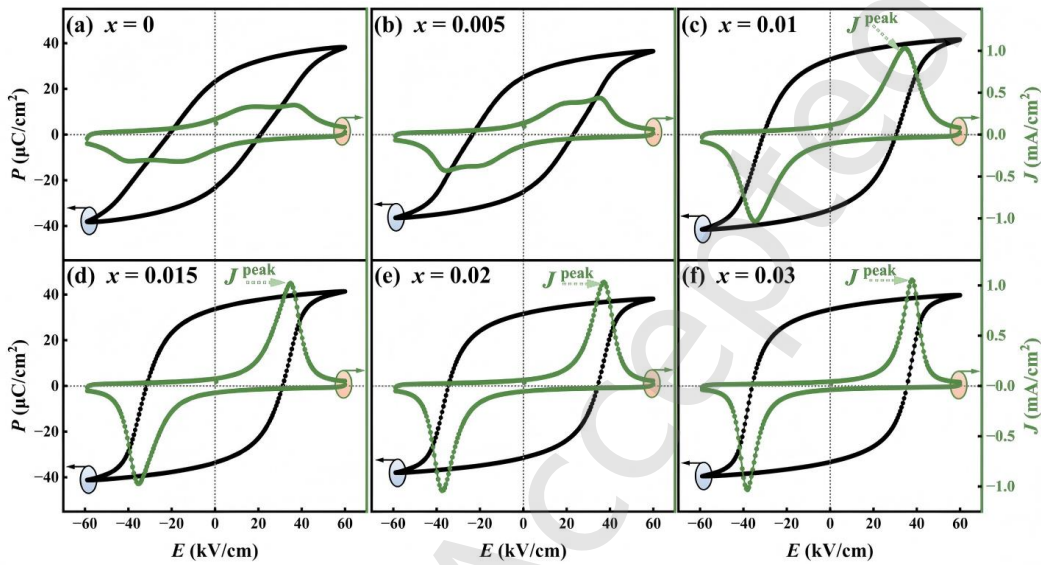


Fig. 3. The P - E loop and J - E curves of BNKBT-Fe(x) piezoceramics.

The electric-field-induced strain response of doped lead-free piezoceramics is generally governed by the combined effects of domain switching, phase evolution, and site-selective defects [37,38]. Fig. 4(a) shows the bipolar electric-field-induced strain curves (bipolar S - E) of BNKBT-Fe(x) ceramics. All ceramics exhibit the standard butterfly-shaped strain curves with obvious negative strain (S_{neg}). The magnitude of S_{neg} first increases and then decreases with increasing x , reaching a maximum value of -0.11% ($x = 0.01$). This result indicates a relatively pronounced contribution from field-induced non- 180° domain switching. The positive strain (S_{pos}) initially decreases and then increases with increasing x , reaching a minimum value of 0.14% ($x = 0.015$). In addition, when $x \geq 0.015$, the overall strain amplitude decreases, which is consistent with the increasing restriction of non- 180° domain switching by oxygen-vacancy-related defect pinning [18,39]. The unipolar electric-field-induced strain curves

(unipolar S - E) of BNKBT-Fe(x) ceramics measured under 60 kV/cm are shown in Fig. 4(b). As x increases from 0 to 0.03, the maximum unipolar strain ($S_{\max}$) gradually decreases. The undoped ceramic ($x = 0$) exhibits the largest $S_{\max}$ with relatively obvious strain hysteresis. With increasing Fe content, the unipolar S - E loop becomes narrower, the strain hysteresis decreases, and the linearity between strain and electric field improves. These changes indicate the progressive suppression of large-field nonlinear strain contributions and a hardening-like electromechanical response associated with acceptor-defect-related pinning [20,39,40].

Fig. 4(c) summarizes the variations in the piezoelectric coefficient (d_{33}) and the inverse piezoelectric coefficient (d_{33}^*) of BNKBT-Fe(x) ceramics. The d_{33}^* , calculated as $S_{\max}/E_{\max}$, gradually decreases from 340 pm/V ($x = 0$) to 210 pm/V ($x = 0.03$), consistent with the variation in $S_{\max}$. In contrast, d_{33} initially increases, reaches a maximum value of 180 pC/N ($x = 0.01$), and then decreases to 92 pC/N ($x = 0.03$). The different trends of d_{33} and d_{33}^* originate from their distinct excitation conditions and response mechanisms. The quasi-static d_{33} , measured after poling under a relatively weak dynamic stress, characterizes the direct piezoelectric response around the remanent poled state. In contrast, d_{33}^* , derived from the unipolar S - E curve under a large electric field, contains substantial nonlinear contributions from non-180° domain switching, domain-wall motion, and field-induced structural responses [41]. Combined with the Rietveld refinement and PFM results, the enhanced d_{33} at ($x = 0.01$) may be attributed to a favorable balance between ($R3c/P4bm$) phase-structure modification and defect-related pinning [31,42]. At moderate Fe content, the beneficial phase-structure effect may partially compensate for defect pinning, whereas further Fe addition causes the pinning effect to become increasingly influential and reduces the piezoelectric response [39,40]. High-resolution O 1s XPS spectra were deconvoluted into lattice oxygen (O_L) and defect-related non-lattice oxygen (O_D), as displayed in Fig. S5. Both the peak intensity and relative fraction of O_D gradually increase with Fe doping, revealing the growth of oxygen-related defects. Fig. 4(d) shows the electromechanical coupling factors of BNKBT-Fe(x) piezoceramics under different vibration modes. As x increases to

0.01, thickness-mode electromechanical coupling factor (k_t) increases markedly to 0.42 and subsequently remains at a relatively high level, whereas the radial electromechanical coupling factor (k_p) changes only gradually. Consequently, k_t/k_p reaches its maximum value of 3.22 at $x=0.01$, indicating the most pronounced thickness-mode relative to radial-mode electromechanical coupling.

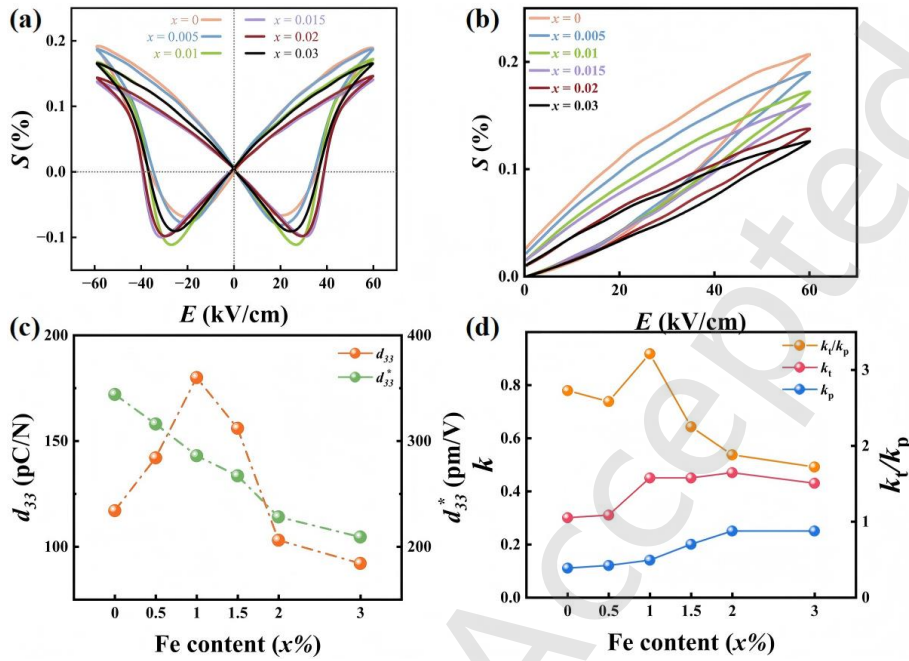


Fig. 4. Strain and electromechanical properties of BNKBT-Fe(x) piezoceramics. (a) Bipolar and (b) unipolar electric field-induced strain curves; (c) variation in the d_{33} and d_{33}^* ; and (d) variation in the k_t , k_p , and k_t/k_p .

3.3 Ultrasonic transducer based on BNKBT-Fe(0.01) ceramic

Compared with the undoped 0.84BNT-0.133BKT-0.027BT ceramics located near the MPB (BNKBT-Fe(0)), the BNKBT-Fe(0.01) ceramic exhibits a higher d_{33} and a larger k_t/k_p ratio, indicating an enhanced electromechanical response with relatively prominent thickness-mode coupling. This indicates that the ferroelectric and piezoelectric properties of BNT-based ternary piezoceramic located at the MPB can be further improved through ion doping modification [43]. Therefore, to verify whether BNKBT-Fe(0.01) can improve ultrasonic transducer performance, a lead-free ultrasonic transducer based on BNKBT-Fe(0.01) piezoceramic with the f_c of 10 MHz was designed and fabricated using the

same electrical matching, acoustic matching, and acoustic beam strategies as in previous work.

Numerical modelling and multilayer/composite structural optimization have been widely employed to improve the acoustic performance of ultrasonic transducers [44,45]. Table S2 lists the material parameters of BNKBT-Fe(0.01) ceramic and other functional layers used for the simulation design in PiezoCAD. By optimizing the thickness and diameter parameters of transducer, the final design parameters were determined, as shown in Table S3. The optical photograph of the fabricated transducer and the schematic illustration of ultrasonic transducer structure are shown in Fig. S6(a-b), respectively. The pulse-echo, tungsten-wire scanning, and porcine-eyeball imaging experiments were performed according to the experimental setups shown in Fig. S6(c-d).

Fig. 5(a-b) shows the simulated impedance-phase spectrum and pulse-echo response of the BNKBT-Fe(0.01) transducer, respectively. At the designed center frequency (f_c) of 10 MHz, the simulated impedance was approximately 50 Ω , with an echo amplitude of 3.16 mV/V and a -6 dB bandwidth of approximately 55%. As shown in Fig. 5(c-d), the fabricated transducer exhibited an impedance of approximately 50 Ω near the f_c , with two resonance peaks at 8.1 and 12.8 MHz. These two peaks may originate from the splitting of the thickness-mode resonance caused by acoustic coupling among the piezoelectric layer, matching layer, and backing layer under the actual device boundary conditions. The measured lower and upper -6 dB frequencies were 7.8 and 13.6 MHz, respectively, corresponding to a f_c of 10.7 MHz and a -6 dB bandwidth of 54%, which agrees well with the simulated value. The differences in the impedance-phase profile and pulse-echo waveform mainly arise from the ideal assumptions of the one-dimensional KLM model, whereas the experimental response is additionally affected by matching-layer loading, bonding interfaces, fabrication deviations, acoustic losses, and lateral vibration modes.

To clarify the contribution of the material properties to transducer performance, Fig. 5(e) compares the insertion loss ($|IL|$) of the BNKBT-Fe(0) and BNKBT-Fe(0.01) transducers. Compared with the BNKBT-Fe(0) transducer, the BNKBT-Fe(0.01) transducer's $|IL|$ decreased from 23 to 16 dB while

maintaining a broad -6 dB bandwidth of 54%. Moreover, the BNKBT-Fe(0.01) transducer exhibits a lower overall $|IL|$ over the same frequency range, and the corresponding minimum $|IL|$ is closer to the designed f_c , indicating it has higher echo sensitivity and energy conversion efficiency. These results demonstrate the material-related advantage of Fe-modified BNKBT ceramics for high-frequency ultrasonic transducers. Fig. 5(f) compares the -6 dB fractional bandwidth and $|IL|$ of representative lead-free ultrasonic transducers reported in the literature [46-57]. The planar BNKBT-Fe(0.01) transducer exhibits a competitive combination of a broad bandwidth and low $|IL|$. These results demonstrate the potential of BNKBT-Fe ceramics for lead-free ultrasonic imaging.

After establishing the material-related performance advantage using the planar transducers, an epoxy acoustic lens was subsequently introduced as a device-level strategy to optimize acoustic focusing and imaging quality. For a transducer with a relatively large active aperture, the acoustic lens concentrates the emitted acoustic energy into a smaller focal region, thereby improving the echo sensitivity and lateral resolution, as illustrated in Fig. 5(g). Fig. 5(h) shows the measured pulse-echo response of the transducer. The peak-to-peak echo voltage (V_{pp}) reaches 2168 mV. The measured f_c is 10.72 MHz, and the -6 dB bandwidth is 31%. Compared with the planar transducer, the -6 dB bandwidth decreased after focusing. This reduction may be attributed to the acoustic lens acting as an additional front acoustic load, which alters the acoustic propagation and frequency-response characteristics, thereby narrowing the originally broad and relatively flat frequency response. Fig. 5(i) compares the transmit-receive performance of the focused BNKBT-Fe(0) and BNKBT-Fe(0.01) transducers. The BNKBT-Fe(0.01) transducer exhibits an approximately threefold increase in V_{pp} , although the bandwidth is reduced.

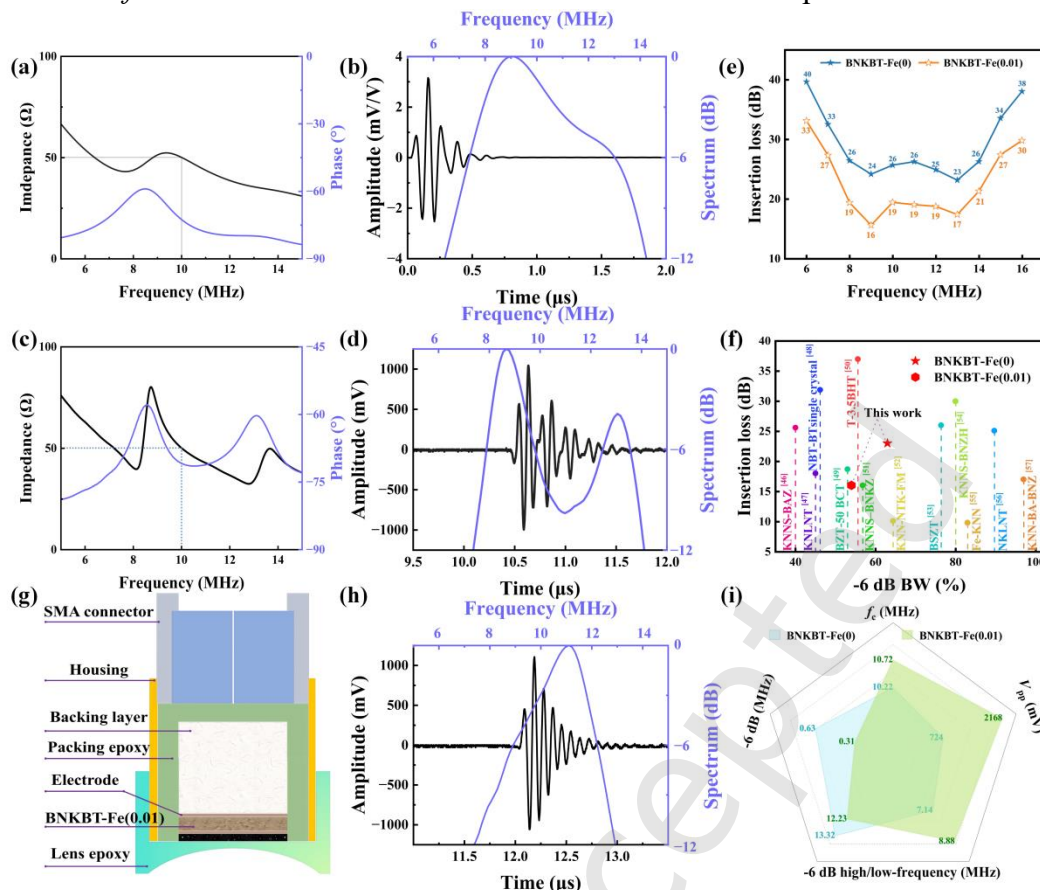


Fig. 5. (a-b) Simulated electrical impedance and phase spectra and pulse-echo response of the planar BNKBT-Fe(0.01) transducer; (c-d) Measured electrical impedance and phase spectra and pulse-echo response of the fabricated planar BNKBT-Fe(0.01) transducer; (e) Comparison of the transmit-receive performance of the planar BNKBT-Fe(0) and BNKBT-Fe(0.01) transducers; (f) Comparison of the -6 dB bandwidth and insertion loss of representative lead-free ultrasonic transducers reported in Refs. [46-57]; (g) Schematic illustration of acoustic focusing using an epoxy acoustic lens; (h) Measured pulse-echo response of the focused BNKBT-Fe(0.01) transducer; (i) Comparison of the transmit-receive performance of the focused BNKBT-Fe(0) and BNKBT-Fe(0.01) transducers.

The imaging resolution of the BNKBT-Fe(0.01) ultrasonic transducer before and after acoustic focusing was tested using an imaging system and a tungsten wire, and the results are shown in Fig. 6. The tungsten wire images acquired using the planar transducer are shown in Fig. 6(a₁-a₃). It is worth noting that the echo signal shows an obvious “ringing effect” in the axial direction, with each wire signal corresponding to multiple parallel bright fringes. This phenomenon can be attributed to the

ceramic. When the same backing layer design strategy is adopted, the backward-propagating acoustic energy may not be sufficiently attenuated, and residual vibration may persist after removal of the excitation pulse, thereby producing a ringing effect. As shown in Fig. 6(a₂-a₃), the axial resolution (R_{axial}) and lateral resolution (R_{lateral}) of the planar transducer are 146 μm and 2834 μm , respectively. Under the same conditions, the wire target was imaged using the focused transducer with an acoustic concave lens, as shown in Fig. 6(b₁-b₃). The focused BNKBT-Fe(0.01) transducer exhibits $R_{\text{axial}} = 182$ μm and $R_{\text{lateral}} = 264$ μm . After focusing, R_{axial} slightly deteriorates, but R_{lateral} is greatly improved. This phenomenon can be attributed to acoustic concave lens focusing, which narrows the acoustic beam near the focal region. However, the relatively short focal length and limited depth of focus may restrict the effective imaging range along the depth direction.

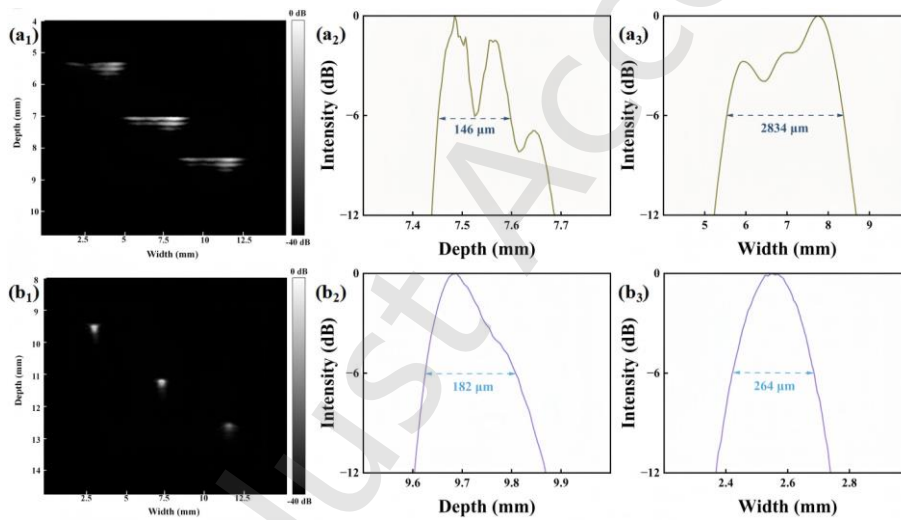


Fig. 6. Resolution measurements of the BNKBT-Fe(0.01) transducer: (a₁-a₃) tungsten-wire image, R_{axial} and R_{lateral} of the planar transducer; (b₁-b₃) corresponding results for the focused transducer.

Fig. 7 shows the imaging results obtained using the BNKBT-Fe(0.01) transducer. Fig. 7(a) shows a schematic cross-section of the porcine eyeball, in which the anterior segment enclosed by the dashed box represents the detection region. The corresponding B-mode image is shown in Fig. 7(b). Based on the difference in acoustic impedance among various tissue layers, distinct grayscale distributions are

observed. The curved hyperechoic band at the top corresponds to the cornea. Below the cornea, the anterior chamber with aqueous humor exhibits high wave transmittance and appears as a dark anechoic area. The hyperechoic structures are the iris and ciliary body, which are located on both sides and below the anterior chamber. The anterior and posterior surfaces of the lens exhibit two curved high-brightness echo bands due to acoustic impedance mismatch, and no reflection signal is observed in the internal region. The vitreous behind the lens has uniform acoustic properties, forming a large anechoic area. Fig. 7(c-d) show the C-mode scanning results of the commemorative coin, where the image grayscale reflects the difference in surface acoustic reflectivity and pattern. The center regions of Fig. 7(c-d) display the “YES” and “NO,” while the peripheral regions show the annular decorative bands and relief patterns. The metal substrate reflects sound waves strongly and produces a bright white hyperechoic background. There is a height difference at the edges of the engraved letters and patterns, which weakens the echo and forms clear black contour lines, thereby showing the details and boundaries of the characters and patterns. Table 1 lists lead-free piezoelectric transducers reported in recent literature.

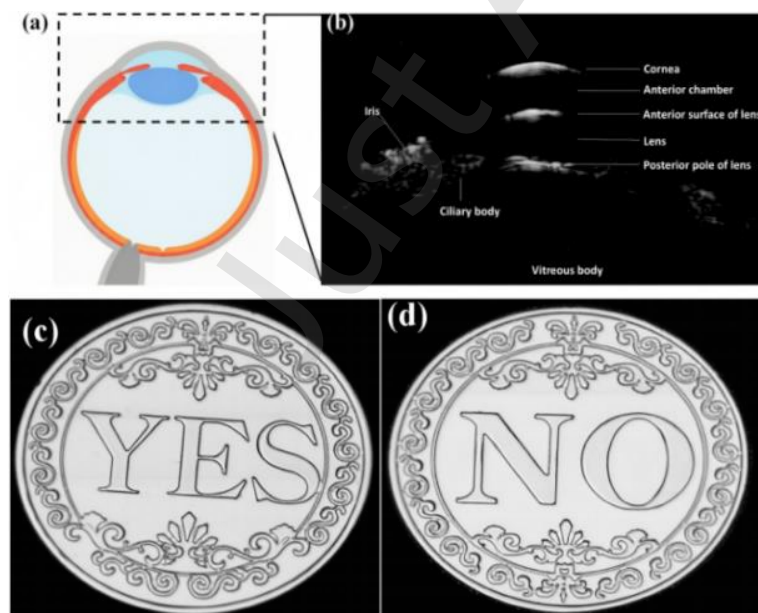


Fig. 7. BNKBT-Fe(0.01) transducer imaging performance: (a) Schematic of the porcine eyeball and (b) B-mode image of the anterior segment; (c-d) C-mode images of the coin.

Table 1. Comparison of representative lead-free and lead-based piezoelectric ultrasonic transducers reported in recent literature.

Material	f_c	-6 dB	$ IL $	R_{axial}	$R_{lateral}$	Imaging	Ref.
	MHz	%	dB	μm	μm		
<i>Lead-free piezoelectric transducers</i>							
BNT-BKT-BT	10	63	23	183	335	wire phantom	[11]
KNLNT	40	45	18	32	102	wire phantom	[47]
BZT-50BCT	30	53	18.7	-		IVUS/coronary artery	[49]
BSZT	40	76.4	26	21.7	95.7	wire phantom	[53]
KNN-based	24.5	97	17	-		porcine eyeball	[57]
BNT-BT	34	26	-	77	484	wire phantom	[58]
BNKBT-Fe	10.72	31	-	182	264	porcine eyeball	This work
<i>Lead-based piezoelectric transducers</i>							
Sm-PMN-PT ceramic	12.4	70	20.5	70	350	Chicken breast	[59]
Sm-PMN-PT single-crystal	15.7	73	14.6	69	545	Porcine belly	[60]
PIN-PMN-PT single-crystal	20	77	19.7	77	125	Tungsten-wire phantom	[61]
PMN-PT free-standing film	82	65	23	35	176	Rabbit aorta/IVUS	[62]

4 Conclusions

This work systematically investigated the effects of B-site Fe^{3+} substitution on the structural evolution and electrical properties of 0.84BNT-0.133BKT-0.027BT lead-free piezoceramics and demonstrated application in ultrasonic transducers. The optimized BNKBT-Fe(0.01) ceramic exhibits $d_{33} = 180$ pC/N and $k_t = 0.42$. Combined with the Rietveld refinement, PFM, and electrical characterization results, the enhanced electromechanical performance is attributed to a favorable balance between ($R3c/P4bm$) phase-structure modification and defect-related pinning. Although Fe^{3+} acceptor substitution introduces oxygen vacancies even at low doping levels, further Fe addition increases the influence of oxygen-vacancy-related defects, thereby restricting domain-wall motion and reducing the

piezoelectric response. Based on this piezoceramic, a 10 MHz transducer was fabricated and achieved good electrical impedance matching. The BNKBT-Fe(0.01) transducer exhibits a higher $V_{pp} = 2168$ mV and a lower $|IL| = 16$ dB. Moreover, the focused transducer achieved $R_{axial} = 182$ and $R_{lateral} = 264$ μm , respectively, providing the spatial resolution required for high-quality imaging. In the imaging application, the BNKBT-Fe(0.01) transducer clearly visualized the internal structures of the porcine eyeball and the surface patterns of the commemorative coin. This study provides a new approach for the development of lead-free ultrasound imaging technology.

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Author contributions

Guangzhi Dong: Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing - review & editing. **Bilin Zhang:** Data curation, Formal analysis, Validation, Visualization, Writing-original draft, Writing - review & editing. **Yifan Zhang:** Formal analysis, Investigation, Validation, Visualization. **Ning Yang:** Data curation, Investigation, Validation, Visualization. **Xinyang Guo:** Formal analysis, Investigation, Validation, Visualization. **Yi Quan:** Conceptualization, Data curation, Methodology, Supervision, Validation. **Chunlong Fei:** Data curation, Formal analysis, Funding acquisition, Supervision. **Yintang Yang:** Methodology, Project administration, Resources. **Haitao Huang:** Methodology, Project administration.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

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