

Complex Analysis of High-Temperature Piezoelectric Ceramics Combining Sr and Nb Co-Doped Bi₄Ti₃O₁₂

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Abstract: The Nb-modified Bi₄Ti₃O₁₂ (BIT + xmol% Nb₂O₅) layer structured piezoelectric ceramic is obtained by the solid-phase method. The study showed that as the content of Nb₂O₅ was increased, the ab-plane-oriented grains were gradually increased, and the grain size became more refined and uniform. The introduction of Nb₂O₅ has significantly reduced the electrical conductivity and dielectric loss of the BIT series ceramics and increased the relative density as well as the piezoelectric and electromechanical properties of ceramics. The results indicate that all samples are single bismuth layer structured phase ceramics. The proper introduction of Sr makes the grain size of BTNO ceramics refined and uniform, presenting dielectric dispersion, and improves its piezoelectric, electromechanical, and ferroelectric properties. When $x = 0.50$, the sample has the optimal performance as the following: relative density $\rho = 98.8\%$, piezoelectric constant $d_{33} = 22$ p C/N, planar electromechanical coupling factor $k_p = 9.5\%$, mechanical quality factor $Q_m = 4462$, remnant polarization $P_r = 13.01$ μ C/cm², Curie temperature $T_c = 62^\circ\text{C}$. In addition, the research on its dielectric properties and thermal stability has suggested that the material ($x = 0.50$) has excellent piezoelectric stability and is suitable for the preparation of high-temperature and high-frequency piezoelectric devices.

Key words: Bi₄Ti₃O₁₂; Electromechanical Properties; Microstructure; Piezoelectric Ceramics

1 INTRODUCTION

The bismuth layer structured ferroelectric (BLSF) material is a lead-free piezoelectric material characterized by a higher Curie temperature (T_c) in the piezoelectric body, and this feature allows it to be applied in extreme environments with high temperature [1-2]. Bi₄Ti₃O₁₂ (BIT) is a typical high Curie temperature bismuth layer structured (BLSFs) material ($T_c \sim 75^\circ\text{C}$). However, due to its low symmetry, spontaneous polarization can only be rotated in two dimensions in the a-b plane, which makes it difficult to obtain sufficient remnant polarization (P_r). In addition, the samples themselves have a low density and are not easy to be fully polarized [8-9], leading to low piezoelectric performance, which further limits its application in instruments and devices [10-11]. In addition to a high T_c , it also has relatively low temperature coefficients of dielectric constant, piezoelectric constant, and resonance frequency, a low aging rate, and a high anisotropic electromechanical coupling factor [3-4], which makes it suitable to be used as materials for pressure sensors, filter devices, and other instruments [5-6]. In addition, its composition does not contain the element Pb, and its anti-fatigue property is also superior to that of traditional lead zirconate titanate (PZT)-based materials, which has attracted extensive attention [7].

In order to improve the electrical properties of BIT materials, massive studies have been carried out both at home and abroad [12-13]. It is found that doping with B-site ions (V^{5+} , Nb^{5+} , W^{6+} , and so on) to modify BIT ceramics can promote sintering and improve its piezoelectric and ferroelectric properties [14]. In addition, the author has found that the introduction of Nb^{5+} can also enhance its electromechanical performance and thermal stability. However, it has been proposed that doping modification at the A site, especially the simultaneously doping at the A and B sites, can improve the electrical properties of piezoelectric materials even more effectively. For example, Sr^{2+} can replace Na^+ and Bi^{3+} at the A site in $Na_{0.5}Bi_{8.5}Ti_7O_{27}$ and $Na_{1.5}Bi_{2.5}Nb_3O_{12}$ ceramics to improve the ferroelectric properties of the system. The co-modified

BIT ceramics at A and B sites (Nd and V) prepared by Nagata et al. have shown excellent electromechanical properties and a low temperature coefficient of the resonance frequency. In addition, the introduction of SrTiO₃ at a proper amount can also significantly improve the electrical properties of $K_{0.5}Na_{0.5}NbO_3$ ceramics. Hence, the author predicts that the modification effect of doping at both A and B sites (such as Sr and Nb) in BIT ceramics is more evident than that of doping at the B site alone. However, there are relatively few reports on the related studies. Therefore, Complex algorithm for high temperature piezoelectric ceramics is prepared using the solid-phase method in this paper, and the effect of doping with Sr on the phase, microstructure, and electrical properties of BNTO ceramics is studied.

2 EXPERIMENT

Complex algorithm for high temperature piezoelectric ceramics is prepared using the traditional solid-phase method. The Bi₂O₃ (98.94%), TiO₂ (99%), Nb₂O₅ (99.5%), and SrCO₃ (99%) are used as raw materials. The materials are weighed based on the corresponding stoichiometric ratio. The prepared raw materials are mixed, pre-fired, crushed, finely ground, granulated, tableted (~12 MPa), compressed, removed of binder, and sintered (1100 ~ 1200 °C, for 4 h). The finished product is fired into a silver electrode, placed in a silicone oil at 80 °C, and subjected to 5 ~ 12.5 kV/mm polarization for 30 min. The product is then allowed to stand for 24 h, and its electrical properties are determined. The surface morphology and phase structure of the sample are analyzed using a scanning electron probe microscope (SEM, Model JSM-6700F, Japan) and XRD (D8 Advance, Bruker axs). The density of the ceramics is tested by using the Archimedes drainage method. The piezoelectric constant d_{33} is measured using a ZJ-3A quasi-static d_{33} measuring instrument. Its dielectric temperature curve at 1 MHz is determined using a precision impedance analyzer (Agilent 4294A). The plane electromechanical coupling factor (k_p), plane frequency constant (N_p), thickness electromechanical coupling factor

(k_t), thickness frequency constant (N_t), and mechanical quality factor (Q_m) are also measured using a precision impedance analyzer (Agilent4294A) based on the resonance and anti-resonance method.

The phase structure and surface morphology of the samples are analyzed by using XRD (D8 Advance, Bruker axs) and a scanning electron probe microscope (SEM, Model JSM-6700F, Japan). The density of ceramics is tested using the Archimedes drainage method. The piezoelectric constant d_{33} is determined using a ZJ-3A quasi-static d_{33} measuring instrument. Its dielectric temperature curve at 100Hz is measured using a precision impedance analyzer (Agilent4294A) and the intelligent control system by Xi'an Jiaotong University. The plane electromechanical coupling factor k_p and the mechanical quality factor Q_m are measured using a precision impedance analyzer (Agilent 4294A) through the resonance and anti-resonance methods. The electrical hysteresis loop of the samples is obtained by the Sawyer-Tower bridge method.

3 RESULTS AND DISCUSSION

3.1 Microstructure Analysis

Fig. 1a and Fig. 1b are the XRD patterns of the 2θ diffraction angles of the BTNO-Sr series samples in the range of $10^\circ \sim 70^\circ$ and $28^\circ \sim 32^\circ$, respectively. From the

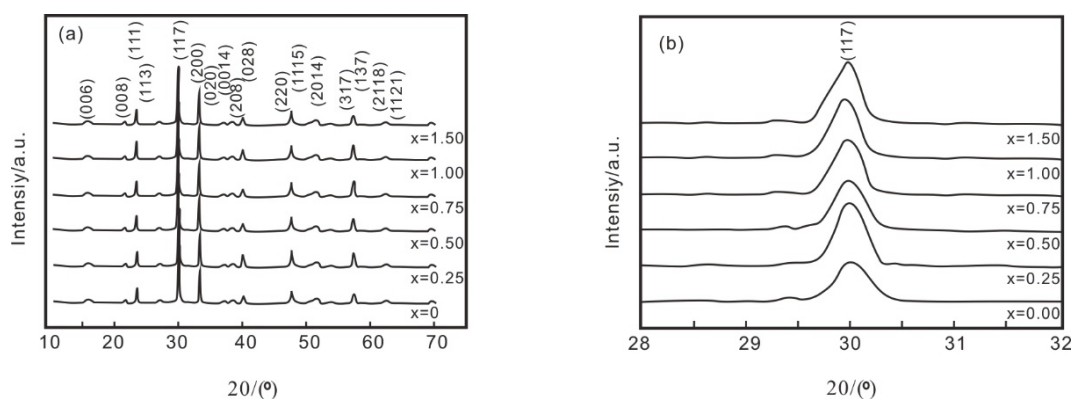


Figure 1 X-ray diffraction (XRD) pattern of the BTNO-Sr powder sample

3.2 Analysis of the Piezoelectric, Dielectric, and Electromechanical Properties

Tab. 1 shows the electrical properties of BTNO-Sr series samples at room temperature. From the table, it can be observed that with the increase of x , the relative density ρ of the sample presents a trend of first decreasing, followed by increasing and then decreasing again. As a result, the d_{33} , Q_m , and k_p also show the same changing pattern. In addition, when $x = 0.50$, the overall performance of the sample is the best, which can reach the maximum values of 98.8%, 22 pC/N, 4462, and 9.5%, respectively. At this point, d_{33} is higher than that of many other bismuth layer structured structure systems. However, Q_m is nearly 13 times that of pure BIT ceramics, even higher than that of NBN single crystal (~ 38) [28]. Hence, it is expected to be applied in the field of high-frequency piezoelectric devices. The ion substitution of different radii and moderate lattice distortion due to vacancies, large polarizability, high P_x (Fig. 2), and so on can all improve

figure, it can be observed that the introduction of Sr did not lead to the appearance of other miscellaneous phases. All samples have an orthorhombic bismuth layer structured phase with $m = 3$, and the crystal face index (117) of the maximum peak is consistent with the maximum peak ($112m + 1$) of the bismuth layer structured ceramics. Fig. 1b shows that the position of the diffraction peak of the sample after doping is slightly shifted to a low angle, which may be due to the addition of Sr and the increase of the A-site ion radius in the ceramic perovskite layer as a result (which is increased from $r_{\text{Bi}^{3+}} = 0.103 \text{ nm}$ to $r_{\text{Sr}^{2+}} = 0.112 \text{ nm}$), leading to fact that the unit cell volume becomes larger. This is consistent with the effect of Sr on the phase of $\text{Na}_{0.5}\text{Bi}_{8.5}\text{Ti}_7\text{O}_{27}$ ceramics. The research has indicated that when the unit cell volume becomes larger [5-6], it is prone to inducing the twisting and deformation of the titanium oxide octahedron, resulting in moderate lattice distortion, which is conducive to the full polarization of the ceramics and the implementation of high electrical properties. In addition, the intensity and width of the diffraction peaks of all samples have shown a very small change, and their positions only have a slight shift. Thus, no significant change has occurred in the theoretical density of the samples, and their relative densities are greater than 97% (as shown in Tab. 1 below). A similar phenomenon has also been observed in the other systems [3-4].

the piezoelectric properties of the sample. Compared with pure BTNO ceramics, the ϵ_r of the doped sample has decreased, and the $\tan\delta$ has increased. When x is increased to 0.50, the ϵ_r and $\tan\delta$ are 213 and 0.74%, respectively. It is reported that the introduction of Sr can also easily make the dielectric properties of the $\text{Na}_{0.5}\text{Bi}_4\text{Ti}_4\text{O}_{15}$ system showing a similar change at room temperature. Therefore, the doping of Sr at a proper amount ($x = 0.50$) can increase the compactness of BTNO ceramics and significantly enhance the piezoelectric and electromechanical properties of the ceramics.

Table 1 Relative density and electrical properties of BTNO-Sr series ceramics

$x(\text{Sr}) / \text{wt}\%$	$\rho / \%$	ϵ_r	$\tan\delta / \%$	$d_{33} / \text{pC}\cdot\text{N}^{-1}$	Q_m	$k_p / \%$
0.00	98.7	224	0.23	18	2804	8.1
0.25	97.2	204	0.56	13	1397	5.4
0.50	98.8	205	0.74	22	4462	9.5
0.75	98.6	207	0.89	18	3072	7.9
1.00	97.3	211	0.92	17	2151	7.1
1.50	97.1	218	1.22	11	1475	3.8

3.3 Analysis of Ferroelectric Performance

Fig. 2 shows the P-E hysteresis loop of a BTNO-Sr ceramic sample. From Fig. 2, it can be seen that the introduction of Sr makes the P-E line of the BTNO sample deviate to the negative axis and present an asymmetric phenomenon, which may be due to the impact of the defects in the crystal structure or the internal bias electric field generated by the motion of the domain wall. It is evident that the shape of the electrical hysteresis loop of the doped sample is more saturated than that of the pure BTNO one. When the amount of Sr added is 0.50, the internal grain size of the ceramic is smaller than that of the pure sample, at ~1 μm only (as shown in Fig. 2 below), and the number of grain boundaries is increased. According to the research, it is necessary to include the polarization reversal in the impact of the grain size at this point; that is, the domain wall is inevitably blocked by the grain boundary during the steering of the electric domain wall. As a result, the E_c of the doped sample is increased to 48.44 kV/cm. On the other hand, the moderate lattice distortion caused by doping is prone to increasing the P_r of the sample, with a value of 13.01 μC/cm², which is higher than that of pure BIT [11]. Hence, the doping with Sr has improved the ferroelectricity of ceramics to a certain extent, increased the polarization rate, and achieved the effect of fully tapping the potential piezoelectric properties, which is conducive to the application of piezoelectric devices.

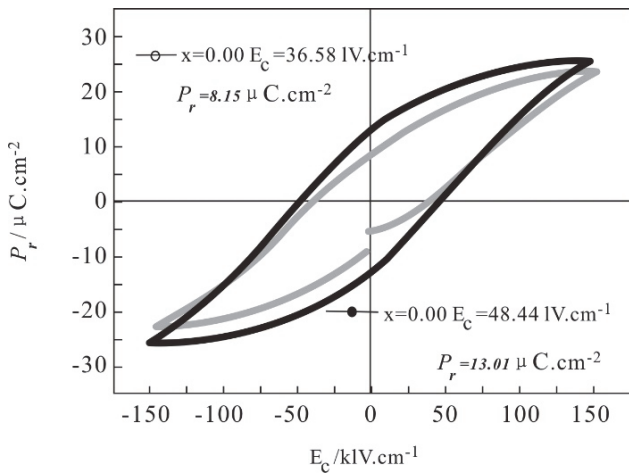


Figure 2 Electric hysteresis loop of BTNO-Sr ceramics

3.4 Analysis of Dielectric Temperature Performance

The BPR optimization matrix can be defined as the following:

$$\psi_{\alpha,s,t}(x) = a^{-3/4} (S_s^{-1} A_\alpha^{-1} (x-t)) \quad (1)$$

In the above equation, $A_\alpha = \begin{pmatrix} a & 0 \\ 0 & \sqrt{a} \end{pmatrix}$, $S_s = \begin{pmatrix} 1 & s \\ 0 & 1 \end{pmatrix}$, which stand for the anisotropic stretching and translation matrix and the shearing matrix, respectively. $\psi_{\alpha,s,t}(x)$ stands for the BPR optimization matrix.

Let $\psi \in L^2(R^2)$, and the following conditions shall be met:

- 1) For $\xi = (\xi_1, \xi_2) \in \hat{R}^2$ ($\xi_2 \neq 0$), $\hat{\psi}(\xi) = \hat{\psi}(\xi_1, \xi_2) = \hat{\psi}_1(\xi_1) \hat{\psi}_2(\xi_2/\xi_1)$ is established, in which $\hat{\psi}(\xi)$ is the Fourier transform of $\psi(\varepsilon)$;
- 2) ψ_1 stands for a continuous wavelet, $\hat{\psi}_1 \in C^\infty(R)$, and $\text{supp} \psi_1 \subset [-2, -1/2] \cup [1/2, 2]$;
- 3) $\hat{\psi}_2 \in C^\infty(R)$, $\text{supp} \psi_2 \subset [-2, -1/2] \cup [1/2, 2]$, in which, $\hat{\psi}_2 \geq 0$ and $\|\hat{\psi}_2\| = 1$ in the interval $(-1, 1)$;

It is assumed that:

$$\sum_{j \geq 0} |\hat{\psi}_1(2^{-2j} \omega)|^2 = 1, \left(|\omega| \geq \frac{1}{8}\right) \quad (2)$$

For each $j \geq 0$, the following is established:

$$\sum_{l=-2^j}^{2^j-1} |\hat{\psi}_2(2^j \omega - l)|^2 = 1, (|\omega| \leq 1) \quad (3)$$

From the support conditions of $\hat{\psi}_1$ and $\hat{\psi}_2$, it can be easily seen that the function $\psi_{j,l,k}$ has frequency support, that is, the following can be obtained:

$$\text{supp} \hat{\psi}_{j,l,k}^{(0)} \subset \left\{ (\xi_1, \xi_2): \xi_1 \in [-2^{2j-1}, -2^{2j-4}] \cup [2^{2j-4}, 2^{2j-1}], \left| \frac{\xi_2}{\xi_1} + l 2^{-j} \right| \leq 2^{-j} \right\} \quad (4)$$

That is, each element $\hat{\psi}_{j,l,k}$ is supported by a pair of trapezoidal structures, the size of which is approximately $2^{2j} \times 2^j$, and its direction is along a straight line at a slope of $l 2^{-j}$.

Fig. 3 shows the dielectric temperature curve of the BTNO-Sr sample at 1 MHz. It is found in the study that with the increase in the ionic radius, the improvement in the symmetry of the crystal structure, and the decrease of the ionic electronegativity, the T_c of the material will be reduced to a certain extent. For the BTNO-Sr samples, when the A-site Bi of the perovskite layer is replaced by Sr, the ionic radius becomes larger. Based on the calculation formula for the tolerance factor t of the perovskite layer, the following can be obtained:

$$t = \frac{R_A + R_O}{\sqrt{2}(R_B + R_O)} \quad (5)$$

In the above equation, R_A , R_B , and R_O stand for the mean radius of A, B, and O ions, respectively. It can be known that the increase in the t of the perovskite layer in the doped sample has led to an increase in the symmetry of the crystal structure. In addition, the electronegativity of the large ion Sr lower than that of Bi and the volatilization of Bi during high-temperature sintering will both lead to a decrease in the T_c of the BTNO-Sr sample. However, it is decreased by 10 °C only and can still remain at 62 °C. In addition, after the introduction of Sr, the A site in the perovskite layer is jointly occupied by Bi and Sr ions, which can easily cause uneven chemical composition and

crystal structure of the sample in the nanoscale. As a result, the normal ferroelectric-paraelectric phase transition temperature is expanded to a phase transition temperature zone, thereby forming microdomain regions with different polarization behaviors in the material structure, in which there are multiple phase transitions in each region as well. Moreover, the bias electric field generated inside the crystal structure mentioned above is also observed. It can induce a dispersion phenomenon in the material, which is manifested as a widening of the dielectric peak of the doped sample at T_c , as shown in Fig. 3 below. It can also be observed from Fig. 3 that when the temperature is lower than 40 °C, the ϵ_r and $\tan\delta$ of the material remain unchanged in essence, which is also reflected in the dense spectrum curve (as shown in Fig. 4 below). When the temperature is further increased, the $\tan\delta$ begins to rise. However, when the temperature rises to 50 °C, the $\tan\delta$ is still at ~5% only. In addition, the variation rate of the dielectric coefficient with the temperature is also relatively low, which suggests that the BTNO-Sr material has excellent dielectric stability.

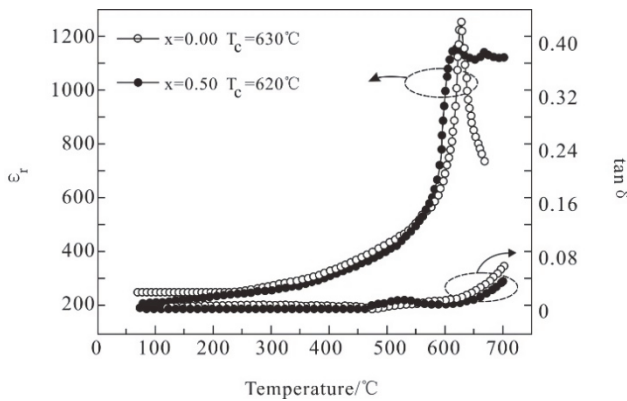


Figure 3 Dielectric temperature curve of BTNO-Sr ceramics

Dense microstructure and uniform grain size will also make .This will be shown as the increase of Nb^{2}O^5 doping . It shows a downward trend. When $x = 4.00$, Minimum 2 orders of magnitude lower than pure bit and 1 ~ 3 orders of magnitude lower than relevant reports [9-10], which is conducive to the smooth high-voltage polarization process of the sample, so as to significantly improve its electrical properties.

From the previous section, it can be known that the piezoelectric, electromechanical, and ferroelectric properties of the sample ($x = 0.50$) at room temperature are the best. As the Curie temperature is high, the high temperature dielectric properties of the sample are further studied. Fig. 4 and Fig. 5 show the relationship between the dielectric constant of BTNO and the doped sample ($x = 0.50$) with the frequency at varying temperatures. It is apparent that when the temperature rises to about 40 °C, the ϵ_r of the sample presents a sharp drop as the frequency is increased, which may be due to the impact of ion migration, so that the $\tan\delta$ of the sample is increased as the temperature in the high temperature region rises (Fig. 3 above). However, when the temperature is lower than 40 °C, the ϵ_r of all samples show a relatively minor change with the increase in the temperature and frequency, and the distribution of the spectral curve is relatively dense and stable. Bit series ceramics were prepared by solid state

method and ordinary molding process (pressure ~ 12 MPa). The effects of Nb^{2}O^5 doping on the phase, microstructure, conductivity, high temperature piezoelectric and electromechanical properties of bit ceramics were studied. As the temperature continues to rise, the distribution of the spectral curve of all samples becomes sparser and sparser gradually. However, the ϵ_r variation amplitude of the doped sample is close to that of the pure BTNO sample (Fig. 5 below). This suggests that the introduction of Sr at a proper amount ($x = 0.50$) can help the BTNO material maintain the dielectric temperature and frequency stability more effectively.

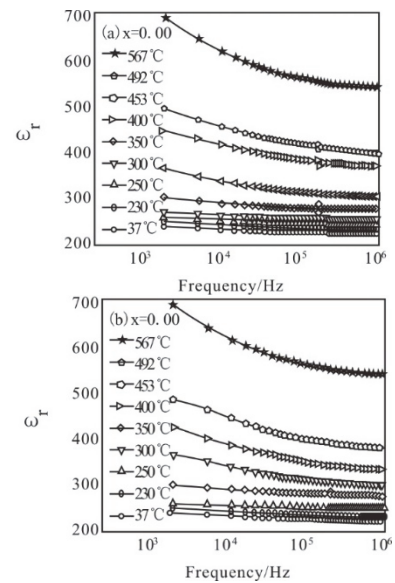


Figure 4 Variation curve for the dielectric constant of BTNO-Sr ceramic samples with the frequency at different temperatures

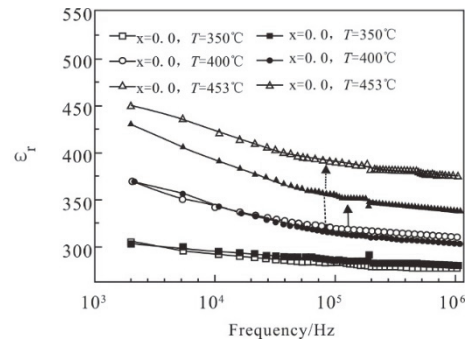


Figure 5 Variation curve for the dielectric constant of BTNO-Sr ceramic samples with the frequency in the temperature range from 35 to 45 °C

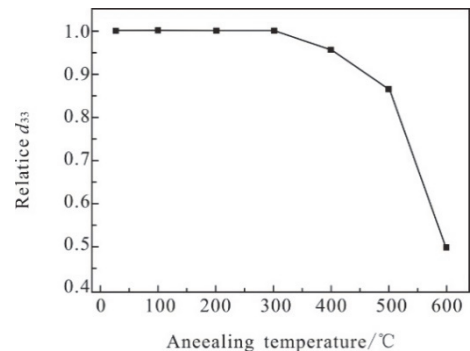


Figure 6 Variation curve for the relative d_{33} of BTNO-Sr ($x = 0.50$) ceramic with the annealing temperature

3.5 Study of the Temperature Stability

The performance of high-temperature piezoelectricity plays an essential role in the proper application of ceramics in piezoelectric devices. In this work, the high-temperature piezoelectric performance of samples doped with Sr was studied, as shown in Fig. 7. The d_{33} of the doped sample is relatively stable with the increase of the annealing temperature, and its change is still not significant at 40 °C. It starts to drop sharply only at 50 °C, and it still has piezoelectric property when it is close to T_c . Through previous work, it is found that the relative d_{33} of BTNO can reach more than 90% when the annealing temperature is 50 °C. Although the relative d_{33} is slightly reduced after the introduction of Sr, it can still be maintained at above 85%. Compared with the other bismuth layer structured systems (such as SrBi₂Nb₂O₉, $T_c \sim 45$ °C), it still has relatively good thermal stability. In addition, the sample has sound electrical properties and dielectric stability at room temperature, so it is suitable to be applied in high-temperature devices.

4 CONCLUSIONS

All samples have a single orthogonal bismuth layer structured structure phase, and the position of their diffraction peak is slightly shifted to a low angle. As a result, there is a moderate distortion of the crystal structure, which is conducive to the improvement of piezoelectric performance. The doping with Sr can improve the compactness and ferroelectricity of ceramics, allow it to be fully polarized, and lead to a significant increase in its electrical properties. In addition, the d_{33} and Q_m of the optimal component $x = 0.50$ are 22 pC/N and 4462, respectively; the introduction of Sr at an appropriate amount ($x = 0.50$) has improved the piezoelectric and ferroelectric properties of BTNO ceramics and significantly enhanced its electromechanical properties so that its T_c is kept at above 600 °C. Its dielectric peak is broadened, presenting dispersibility. In addition, the excellent dielectric and piezoelectric stability of the material are maintained, which indicates that this ceramic component is expected to be one of the promising candidates for high-temperature and high-frequency piezoelectric materials.

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