Influence of component content on the properties of PSZN-PSNN-PSZT ceramics

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Abstract
In this paper, (Pb0.9Sr0.1)(Zn1/3Nb2/3)x(0.06+y(Ni1/3Nb2/3)x)0.16(Zr0.52Ti0.48)0.78-x)O3 (PSZN-PSNN-PSZT, x = 0, 0.02, 0.04, 0.06, 0.08) piezoelectric ceramics were prepared by traditional solid-state reaction method. The samples were studied on microstructure, phase structure, electrical properties and dielectric relaxation. All as-prepared PSZN-PSNN-PSZT ceramics present single phase of perovskite structure. Experimental results show that the variation trend of the R phase percentage content is consistent with the trend of d33. Also, increasing PSZN content will inhibit the growth of sample grains. The temperature-dependent of dielectric performance testing results present that all the samples possess obvious relaxation behavior. The best overall performance of the samples are obtained at x = 0.06, where its d33 = 920pC/N, k33 = 0.71, P r = 24.54 μC cm−2, γ = 1.98, ε r = 6856, tanδ = 3.3%, Tc = 133 °C. The PSZN-PSNN-PSZT ceramics are potential candidate materials due to its simple preparation process, the high dielectric and piezoelectric properties.

1. Introduction
Since the discovery and later application of piezoelectric phenomenon, lead-based ceramics have attracted widespread attention due to their excellent piezoelectric, dielectric properties [1, 2]. Among them, lead zirconate titanate (PZT) is the focus of research [3–6]. With the development of science and technology, the performance of piezoelectric ceramics urgently needs to be improved. Pure PZT is difficult to meet the demand of piezoelectric ceramic products in terms of property. Therefore, doping metal elements and introducing components have become important methods in exploring new formulations [7, 8]. After the unremitting efforts of the researchers, the multi-component piezoelectric ceramic formula is now widely used for its excellent electrical properties [9–11]. Nowadays, the research on multi-components mainly includes methods such as process optimization, doping of metal elements, component control and component introduction [7, 8]. In particular, component control is a more feasible method for multiple components. The interaction between the components is relatively large, which affects the performance of the ceramic. Therefore, it is necessary to optimize the performance of a multi-component formulation by component control. Vittayakorn et al found the MPB composition point of (1-x)PNN-xPZT ceramics through component control [12]. Li et al increased the piezoelectric constant and electromechanical coupling coefficient of 0.28PZN-0.72PZT ceramic through component control [13]. PZN-PNN-PZT piezoelectric ceramics have well electrical properties, so certain research [9, 14, 15]. Peng et al improved the properties of PZN-PNN-PZT piezoelectric ceramics by adjusting the PNN/PZN ratio, with d33 = 800pC/N, k33 = 0.63, ε r = 6020 [7]. Zhang et al researched the effect on the electrical properties of PZN-PNN-PZT by adjusting PZN/PZT and doping Sr3+1, finding that d33 = 800pC/N, k33 = 0.65, ε r = 4081 [16]. So far, the d33 and k33 of reported PZN-PNN-PZT piezoelectric ceramics are not very high. It is especially important to improve the electrical performance by adjusting the component of the multi-component piezoelectric ceramic formulation. In this paper, the microstructure, phase transition structure, electrical properties and dielectric relaxation behavior of PSZN-PSNN-PSZT piezoelectric ceramics are studied by adjusting the ratio of PSZN/PSZT. The results show that adjusting the ratio of PSZN/PSZT effectively...
improves the electrical properties of PSZN-PSNN-PSZT piezoelectric ceramics, to a higher level that $d_{33} = 920 \mu C/N$, $k_p = 0.71$, $P_r = 24.54 \mu C \ cm^{-2}$, $\gamma = 1.98$, $\varepsilon_r = 6856$, $\tan \delta = 3.3\%$, $T_c = 133 \ ^\circ C$. As a result, PSZN-PSNN-PSZT piezoelectric ceramics have the potential to be used as low-heat and high-amplitude actuators.

2. Experimental method

The (Pb$_0.9$Sr$_{0.1}$)(Zn$_{1/3}$Nb$_{2/3}$)$_x$(Ni$_{1/3}$Nb$_{2/3}$)$_{0.16}$-(Zr$_{0.52}$Ti$_{0.48}$)$_{0.16}$O$_{2}$ ceramics were prepared by the traditional solid-state reaction method. Mix Pb$_2$O$_4$ (99%), SrCO$_3$ (99%), ZnO (99%), Nb$_2$O$_5$ (99%), Ni$_2$O$_3$ (98%), ZrO$_2$ (99%) and TiO$_2$ (99%) raw materials in stoichiometric ratio, where the used materials are analytical reagent. In order to prevent Pb from volatilizing during sintering, Pb$_3$O$_4$ should be a 0.5wt% excess when weighing. The mixed raw materials were ball milled in stoichiometric ratio, where the used materials are analytical reagent. In order to prevent Pb from volatilizing during sintering, Pb$_3$O$_4$ should be a 0.5wt% excess when weighing. The mixed raw materials were ball milled in ethanol medium for 12h, then calcined it under 900$^\circ\ C$ for 4h after dried. The calcination powder was subjected to secondary ball milling under the same conditions for 14h. Mixing The dried powder with 5wt% paraffin uniformly, and under 8MP it was pressed into small discs with diameter of 12mm and a thickness of 1mm. Finally, the sample was put into a closed sintering furnace at 1260$^\circ\ C$ for 2h, with the heating rate of 2$^\circ\ C$/min, and then cooled to room temperature along with the furnace.

X-ray diffractometer (XRD, Model PANalytical/X’pert-PRO, Holland) was used to test the crystal phase of the ceramic sample. The surface morphology of the ceramic samples was analyzed by scanning electron microscope (SEM, Model ZEISS MERLIN Compact, America). The surface of the sample was brushed with silver electrode. It was baked under 500$^\circ\ C$ for 30 min, then cooled to room temperature. It was next polarized in a 2.5 kV mm$^{-1}$ DC electric field and a 50$^\circ\ C$ silicone oil bath for 30 min Measure the piezoelectric constant $d_{33}$ of ceramic samples with a quasi-static $d_{33}$ tester (ZJ-3A, Institute of Acoustics, Chinese Academy of Sciences, China). Capacitance (C) and dielectric loss (tan$\delta$) were measured by capacitance terse (Model TH2618B) at room temperature under 1kHz, next the dielectric constant ($\varepsilon_r$) was calculated with capacitance (C). The change of relative dielectric constant and dielectric loss with temperature was tested by the LCR meat (Agilent, E4980A) which is connected to a computer-controlled greenhouse, and the frequency ranges from 0.1kHz to 100kHz. The electromechanical coupling coefficient ($k_p$) is calculated by testing resonance and anti-resonance with an impedance analyzer (Agilent 4294A). According to the follow formula [8].

$$\varepsilon_r = \frac{\varepsilon}{\varepsilon_0} = \frac{Cd}{S\varepsilon_0} = \frac{Cd}{\varepsilon_0 \pi \Phi^2} \approx \frac{144Cd}{\Phi^2}$$

$$kp = \sqrt{2.45 \times \left(\frac{f_a - fr}{fr}\right)}$$

The free space dielectric constant value $\varepsilon_0 = 8.85 \times 10^{-12} \ F \ m^{-1}$, C$^{-1}$ represents capacitance (units $10^{-12} \ F$), $d$ means the sample thickness (mm), $S$ denote the area (mm$^2$), $\Phi$ is the sample diameter, $f_r$ is the resonance frequency, $f_a$ is the anti-resonance frequency.

3. Experimental results and analysis

3.1. Phase structure of PSZN-PSNN-PSZT ceramics

XRD patterns of the samples and corresponding refined results by Rietveld software are shown in figure 1(a)-(e). It can be seen that all ceramics possessed pure perovskite structure and only R-T phase coexistence was observed in the samples. However, as $x$ increases, the proportion of T phase and R phase happened to changes. The content of T phase decreases with the increase of $x$, while the content of R phase shows the opposite trend. When $x = 0.04$ and 0.06, the two-phase content is almost unchanged. When $x$ further increases to 0.08, the T phase content increases and R phase content decreases. For investigation of lattice change induced by the PSZN content, lattice parameters of T phase and R phase are presented figure 1(f)-(h). The lattice parameters $a$ of the T phase gradually decreases with the increase of $x$, and the lattice parameters $c$ first decreases, then increases, and finally decreases. For the R phase, with the increase of $x$, a first increases sharply and then remains almost unchanged. The $c/a$ of the T phase decreases first, then increases, and finally decreases gradually. By checking relevant information and analyzing the proportion of ZN and ZT in the formulation, the average radius of Zn$^{2+}$ and Nb$^{5+}$ are 0.068 nm (0.074/3 + 0.064 × 2/3), and that of Zr$^{4+}$ and Ti$^{4+}$ are 0.063 nm, which means that the increase of $x$ will lead to the increase of lattice constant and lattice distortion. Wang et al found in the KNN system that doping at sites A and B has no direct effect on the crystal lattice parameters [1]. The lattice distortion may be due to the overlap of electron orbits among various ions in the same sites, which led to the restriction through electrostatic forces [17]. Besides, orbital polarization was verified to be equal with spin polarization and...
charge density for strongly interacting electron systems, so orbital ordering was also considered to be of great importance for crystal structure and parameters of the exchange interaction [18, 19]. All lattice parameters of the samples are listed in table 1.

### 3.2. Surface microstructure of ceramic

Figure 2 shows that the cross-sectional micro-morphology of (0.9P0.1S)\((0.06+x)\)ZN-0.16NN-\((0.78-x)\)ZT ceramics was sintered at 1260°C for 2h. Sufficient grain growth is obtained in all the samples at the present sintering conditions and not too many pores existed in the samples. To better compare the grain size for different contents of PSZN, the distribution of grain size and the average grain size were analyzed, as shown in figure 3. It is clearly observed that all the samples have relatively homogeneous grain sizes, and the average grain size slowly decreases from 3.34 to 3.14 μm when \(x\) is increased from 0.02 to 0.06; after that, the grain size sharply decreases to 2.64 μm at \(x = 0.08\). This means that PSZN could inhibit grain growth. The electrical properties of piezoelectric ceramics depend to a great extent on the difficulty of electric domain switching, and the electric domain switching depends on the growth of crystal grains. The finer the grains, the greater the number of grain

| Table 1. Rietveld-refinement parameters of (0.9P0.1S)\((0.06+x)\)ZN-0.16NN-\((0.78-x)\)ZT ceramics. |
|-----------------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| \(x\)       | SG      | \(a(\text{Å})\) | \(b(\text{Å})\) | \(c(\text{Å})\) | \(\alpha = \beta = \gamma (^\circ)\) | Fraction (\%) | Volume (Å\(^3\)) | \(\chi^2\) | Rp (%) | wRp (%) |
| 0            | P4mm    | 4.0475  | 4.0475  | 4.0733  | 90   | 80.124   | 66.729 | 1.3     | 5.78   | 7.91    |
| 0.02         | R3m     | 4.0619  | 4.0619  | 4.0619  | 89.894  | 19.876   | 67.019 | 3.7     | 8.48   | 12.90   |
| 0.04         | P4mm    | 4.0442  | 4.0442  | 4.0758  | 90   | 38.709   | 66.663 | 3.4     |        |        |
| 0.06         | R3m     | 4.0580  | 4.0580  | 4.0580  | 89.926  | 61.291   | 66.823 | 3.9     | 9.15   | 12.82   |
| 0.08         | P4mm    | 4.0413  | 4.0413  | 4.0852  | 90   | 14.205   | 66.720 | 3.0     |        |        |
| 0.08         | R3m     | 4.0610  | 4.0610  | 4.0610  | 89.906  | 85.795   | 66.973 | 9.13    | 12.83  |        |
| 0.08         | P4mm    | 4.0404  | 4.0404  | 4.0835  | 90   | 14.040   | 66.664 | 8.55    | 11.66  |        |
| 0.08         | R3m     | 4.0578  | 4.0578  | 4.0578  | 89.915  | 85.960   | 66.814 | 8.55    | 11.66  |        |
| 0.08         | P4mm    | 4.0386  | 4.0386  | 4.0704  | 90   | 38.545   | 66.389 | 9.15    | 12.82  |        |
| 0.08         | R3m     | 4.0541  | 4.0541  | 4.0541  | 89.915  | 61.455   | 66.630 | 9.15    | 12.82  |        |
boundaries. This will lead to increased resistance to the switching of the ceramic internal electric domains during polarization, which will affect the electrical properties of the ceramic.

### 3.3. Electrical performance analysis

It can be seen from figure 4 that as $x$ increases, the $d_{33}$ of the ceramic sample at room temperature first increases and then decreases, but $k_p$ is barely changing, and $\varepsilon_r$ and $\tan\delta$ gradually increase. The macroscopic properties of ceramics are related to the microstructure, combined with XRD pattern and SEM image analysis. The change trend of $d_{33}$ is consistent with the change trend of R phase content in table 1. The increase of PSZN content smaller grains and numerous grains boundaries contributed to pinning effect which effectively inhibited the reversal of domains switching. Thus, excellent dielectric properties were maintained. SEM analysis results show that the grains of all samples are effective growth and the grain distribution is evenly; PSZN-PSNN-PSZT ceramics show excellent comprehensive properties. Generally, smaller grain size corresponds to higher grain boundary volume, and the reduction in grain size may also lead to an increase in dielectric loss. The increase of
4. Dielectric properties of ceramics

The temperature dependences of dielectric constant and dielectric loss measured at frequencies of 0.1, 1, 10 and 100 kHz for the 1260°C-sintered (0.9P0.1S)(0.06+x)ZN-0.16NN-(0.78-x)ZT ceramics with x = 0, 0.02, 0.04, 0.06 and 0.08 are shown in figure 5. It can be clearly seen from figure 5 that all samples have a broad dielectric peak, this phenomenon is called a diffuse phase transition. In addition, as the frequency increases, the phase transition temperature gradually shifts to the right, and the maximum dielectric constant gradually decreases, this phenomenon is called frequency dispersion. This are two important characteristics of relaxor-type ferroelectrics [20]; According to previous reports, the relaxation behavior of ceramics is related to polar nano-domains. In this study, Zn2+ and Nb5+ have different radii and valences from Zr4+ and Ti4+ ions. Replacing Zr4+ and Ti4+ with Zn2+ and Nb5+ will cause local charge balance and lattice distortion. This will lead to the PSZN-PSNN-PSZT ceramics to produce polar nano-domains that result relaxation behavior [20]. Therefore, it can be concluded that the prepared ceramic is a relaxor-type ferroelectric material. It can be seen from table 2 that tanδ (max) increases first and then decreases with the increase of x. The dielectric properties of the sample are higher than other samples at x = 0.06, which may be due to the decrease of the non-polar region of the ceramic induced by the phase transition [16]. Tc decreases with the increase of x, which may be caused by: On the one hand, because the Curie temperatures of PZN, PNN, and PZT (Zr/Ti = 0.52/0.48) are 140 °C, −120 °C, and 350 °C respectively, so with the increase of x the Tc of (0.90.1S)(0.06+x)ZN-0.16NN-(0.78-x)ZT ceramic will decrease [16]. On the other hand, it may be caused by the volatilization of Zn [9]. It can be that he PZN composition would decompose into cubic pyrochlore, ZnO and PbO at high temperature [21, 22]. When the
sintering temperature rises to 800–900°C, PbO and ZnO form a low melting point eutectic, which leads to the volatilization of PbO and ZnO, which leads to a decrease in the $T_c$ of PSZN–PSNN–PSZT ceramics.

### 3.5. Relaxation behavior of ceramics

Figure 6 shows the relationship between $1/\varepsilon$ of (P0.9P0.1S)[(0.06+x)ZN-0.16NN-0.87-x)ZT] ceramic sample and temperature ($T$) at 10 kHz. For normal ferroelectrics, when the temperature is higher than the Curie temperature, the dielectric permittivity should follow the Curie-Weiss law described by references [8];

$$\frac{1}{\varepsilon} = \frac{T - TCW}{C}$$

Where, $C$ is the Curie-Weiss constant; $\varepsilon$ is the dielectric constant of the sample; $T$ is the temperature of the ceramic sample; $TCW$ is the Curie-Weiss temperature. $TCW$ for different components can be obtained by formula fitting (3). As can be seen from figure 6, the level that the relationship between $1/\varepsilon$ and $T$ deviates from the Curie-Weiss law around the Curie temperature of each sample can be defined by the formula [8];

$$\Delta T_m = T_B - T_m$$

Where, $T_B$ is the initial temperature where the dielectric constant conforms to Curie’s law; $T_m$ is the temperature corresponding to the maximum dielectric constant of the ceramic. It can be seen from figure 6 that the dielectric constant of the ceramic sample deviates from the Curie-Weiss law near the Curie temperature, and it does not begin to meet the requirement until the temperature rises to $T_B$. In figure 6. There is an anomaly in permittivity temperature dependence, at ~100 °C, for $x = 0.00$ and 0.06. This may be because at this composition where has a T–phase and R-phase transition in this temperature range. At low temperature, the space charge or defect dipole in the ceramic sample is frozen at a relatively stable lattice potential energy position, showing a long relaxation time and weak frequency dependence. When the temperature rises, the thermal motion of the space charge or defect dipole intensifies and is no longer frozen. At this time, the relaxation time is reduced, showing a clear frequency dependence; When the temperature further rises, the relaxation time is further reduced, the frequency dependence is weakened. Table 3 shows some values about dielectric properties. From the table 3, there is no obvious change about $\Delta T_m$ with increase of $x$ content. It indicates that the dielectric relaxation of all samples is relatively close.

For relaxor ferroelectrics, we can study them according to the modified Curie-Weiss law [8];

$$\frac{1}{\varepsilon} - \frac{1}{\varepsilon_m} = \frac{(T - T_m)^\gamma}{C_1} \quad (T > T_m, 1 \leq \gamma \leq 2)$$

where $C_1$ is the Curie constant, $\varepsilon_m$ and $T_m$ are the maximum dielectric constant and temperature correspondingly, $\gamma$ is called a diffusion coefficient ranging from 1 (normal ferroelectric) to 2 (complete relaxor ferroelectric).

Figures 7(a)–(e) show graph of the relationship between $\ln(1/\varepsilon - 1/\varepsilon_m)$ and $\ln(T - T_m)$ at 10 kHz. Figure 7(f) shows the change trend of $\gamma$. The $\gamma$ can be obtained by fitting the experimental data with the formula (5). The dispersion coefficients of all samples have larger values. When $\gamma$ is between 1–2, the sample is relaxor ferroelectric. And the larger $\gamma$ indicates the sample the more obvious the relaxation behavior. When $x = 0.04$ and 0.06, $\gamma = 1.98$, indicating that the relaxation behavior of this sample is more obvious than that of other samples. With the increase of $x$, the dispersion coefficient of the ceramic sample first increases and then decreases; Combined with the XRD analysis results, it can be seen that the change trend of $\gamma$ value is consistent with the change trend of R phase percentage content. This may be due to the better stability of R phase compared with T phase. The reason why may be attributed to that the R phase possesses better stabilities than the T phase. With the increase of PSZN content to 0.06, the enlarged diffuseness coefficient indicates the gradual increase of diffuse phase transition behavior, which can be ascribed to the enhancement of chemical heterogeneities with much more PSZN content. However, when $x = 0.08$, the decrease in $\gamma$ may be caused by the decrease in grain size.

| $x$ | $\varepsilon$ (10kHz max) | $\tan\delta$ (10kHz max) | $T_m$(10kHz) |
|-----|-----------------------|-----------------|-------------|
| 0.00 | 20534                | 0.056           | 154         |
| 0.02 | 16918                | 0.060           | 136         |
| 0.04 | 17264                | 0.064           | 136         |
| 0.06 | 26792                | 0.077           | 133         |
| 0.08 | 19898                | 0.074           | 127         |
3.6. Ferroelectric behavior of ceramics

Figure 8(a) shows the P-E hysteresis loop of \((0.9P0.1S)\[(0.06+x) ZN-0.16NN-(0.78-x)ZT]\) ceramic sample.

Figure 8(b) shows remnant polarization (\(P_r\)) and coercive field (\(E_c\)) of samples. It can be seen from figure 8(a) that the samples exhibit a typical ferroelectric hysteresis loop, and the shape and size of the P-E hysteresis loop are almost unchanged as \(x\) increases. It can be seen from figure 8(b) that as \(x\) increases, \(E_c\) is almost unchanged, \(P_r\) shows a slight downward trend. In addition, the \(P_r\) reduces with the increase of PSZN content,

Table 3. Summary of some parameters extracted from ceramics dielectric measurements at 10 kHz.

| \(x\)  | \(T_{CW}/^\circ C\) | \(T_B/^\circ C\) | \(T_m/^\circ C\) | \(\Delta T_m/^\circ C\) | \(\varepsilon_m\) | \(\gamma\) |
|-------|-------------------|-----------------|-----------------|---------------------|----------------|--------|
| 0.00  | 187               | 228             | 154             | 74                  | 20532          | 1.91   |
| 0.02  | 180               | 217             | 136             | 81                  | 16918          | 1.95   |
| 0.04  | 175               | 220             | 136             | 84                  | 17264          | 1.98   |
| 0.06  | 169               | 207             | 133             | 74                  | 26792          | 1.98   |
| 0.08  | 168               | 209             | 127             | 82                  | 19898          | 1.82   |

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Figure 6. The inverse dielectric constant at 10 kHz as a function of temperature for the \((0.9P0.1S)\[(0.06+x) ZN-0.16NN-(0.78-x)ZT]\) ceramics.

Figure 7. Plot of \(\ln(1/\varepsilon_m) - \ln(1/\varepsilon_m)\) vs. \(\ln(T-T_{m})\) at 10 kHz for \((0.9P0.1S)\[(0.06+x) ZN-0.16NN-(0.78-x)ZT]\) ceramic samples.
which may be caused by the enlargement of lattice distortion and diffuse phase transition behavior. In summary, it is believed that as \( x \) increases, it has little influence on the ferroelectric properties of ceramics.

4. Conclusions

In this paper, traditional solid-phase reaction is used to prepare PSZN-PSNN-PSZT piezoelectric ceramics. The effects of adjusting the ratio of PSZN/PSZT on the phase transition, microstructure, electrical properties and relaxation behavior of ceramics was systematically studied. XRD analysis results show that all samples are of pure perovskite structure, with only the coexistence of T phase and R phase. According to the analysis of SEM results, the grain size distributions of all samples are similar. As the PSZN content increases, the average grain size of the samples decreases. The temperature-dependent of dielectric constant has presented diffuse phase transition and frequency dispersion behavior, the ceramics are a relaxor-type ferroelectric material despite it shows typical ferroelectric hysteresis loop. The increase of PSZN content has almost no effect on the \( P_r \) and \( E_c \) of PSZN-PSNN-PSZT ceramics. When \( x = 0.06 \), the ceramic has the best overall performance; \( d_{33} = 920 \text{pC/N}, k_p = 0.71, P_r = 24.54 \mu \text{C/cm}^2, \gamma = 1.98, \varepsilon_e = 856, \tan \delta = 3.3\%, T_c = 133 \degree \text{C} \). The experimental results show that this PSZN-PSNN-PSZT ceramics have the potential for low-heat and high-amplitude actuators.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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