Composition design of PMN-PH-PT piezoelectric ceramics for high-temperature actuator applications

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Abstract

High-Curie temperature (Tc) piezoelectric ceramics 0.15Pb(Mg1/3Nb2/3)O3-xPbHfO3-(0.85-x)PbTiO3 (0.15PMN-xPH-(0.85-x)PT, x = 0.36, 0.38, 0.39, 0.40, 0.42, 0.15PMN-PH-PT) were synthesized by the solid-state reaction method via the columbite precursor route. The synthesized 0.15PMN-PH-PT ceramics exhibit pure perovskite structure with compositions locate near the morphotropic phase boundary (MPB), where the rhombohedral phase and the tetragonal phase coexist. The sintered samples present high densification, in which the 0.15Pb(Mg1/3Nb2/3)O3-0.38PbHfO3-0.47PbTiO3 (0.15PMN-0.38PH-0.47PT) ceramics present the highest relative density, being 97.76%. The dielectric performance-temperature curves measured at different frequencies, the Curie-Weiss law fitting and the quadratic law fitting indicate that the ferroelectric phase transition from ferroelectric phase to paraelectric phase occurred at T0 in the 0.15PMN-PH-PT ceramics is driven by the displacive phase transition, and the 0.15PMN-PH-PT ceramics belong to normal ferroelectrics with diffused phase transition characteristic. Due to the MPB effect and high relative density, the 0.15PMN-0.38PH-0.47PT ceramics exhibit the best integral electrical properties, excellent thermal stability and large electric field induced strain, which present prospect promising for actuator applications under elevated environmental temperatures.

1. Introduction

In recent decades, electronic and electrical devices used in special environmental put forward severe requirements for piezoelectric materials. Among which detonation sensors used in automobile engines and acceleration sensors used in aerospaces are required to work under high temperatures, and acoustic logging instruments used in oil and gas exploration should tolerance high temperature and high pressure [1–3]. Therefore, developing high Curie temperature (Tc) and high energy density piezoelectrics have become research hotspots [4–7].

The most worldwide commercially used piezoelectrics are lead zirconate titanate (PZT) ceramics and PZT-based solutions nowadays. However, the typical Tc temperature is 193 °C for the commercial PZT5H ceramics, which is relatively low and can not meet the requirements of the above applications [8, 9]. High Tc piezoelectric single crystals, some present excellent piezoelectricity but the Tc is not high enough [10], and some have small piezoelectric constant (d33) although possess high Tc temperature, i.e., 6 pC N−1 and 1210 °C of LiNbO3 [11], whereas their crystal growth is all expensive and difficult, which restricts the applications in electronic and electrical devices. High Tm ceramics such as BiScO3 and Bi(Ni1/2Ti1/2)O3-based system, their preparation
process is also difficult; furthermore, their leakage current is too large due to the evaporation of some elements during sintering, leading to polarization difficulty and limiting their applications [4, 5, 12].

Recently, relatively novel pseudo-ternary Pb(Mg0.33/Nb2/3)O3-PbHfO3-PbTiO3 (PMN-PH-PT) ceramics were reported, which could be prepared easily and exhibited comparable piezoelectricity (piezoelectric constant d33 = 680 pC N⁻¹) with that of Pb(Mg0.33/Nb2/3)O3-PbZrO3-PbTiO3 (PMN-PZT, d33 = 698 pC N⁻¹, Tc = 200 °C), but with rather high Tc; temperature, i.e., 276 °C [13–16]. Such PMN-PH-PT ceramics present promising applications in high-temperature actuators, whereas the study on this system is inadequate. Therefore, in this work, the PMN-PH-PT ceramics with compositions around the morphotropic phase boundary (MPB) were chosen, and the effects of composition design on structure, density and electrical properties were studied.

2. Experimental procedure

The 0.15Pb(Mg0.33/Nb2/3)O3-xPbHfO3-(0.85-x)PbTiO3 (0.15PMN-PH-PT, x = 0.36, 0.38, 0.39, 0.40, 0.42) ceramics were prepared by the solid-state sintering method via the columbite precursor route [17, 18]. Via the two-step synthesis route, columbite precursor MgNb2O6 (MN) was synthesized first, and then reacted with other necessary oxides, which can avoid the direct reaction between Nb2O5 and PbO, inhibit the formation of pyrochlore phase effectively, and promote obtaining perovskite target product [17, 18].

Analytical purity oxides, i.e., MgO (98%), Nb2O5 (99.5%), HfO2 (99.9%), TiO2 (98%) and PbO (99%), were used as raw materials, which were dried fully to ensure weighing stoichiometry. Columbite precursor MN was synthesized by calcining the well mixed oxides of stoichiometric MgO and Nb2O5 at 1000 °C for 2 h. After pulverization and grinding, stoichiometric HfO2, TiO2 and PbO were added, and the homogeneously mixed mixtures were calcined at 800 °C for 4 h to form perovskite 0.15PMN-PH-PT powder. The following ceramics synthesis process was reported elsewhere [19], in which the ceramics were sintered at 1260 °C for 2 h and the Pb-rich sintering atmosphere was provided by PbZrO3, which was prepared by calcining the well mixed stoichiometric PbO and ZrO2 at 850 °C for 4 h.

The sintered 0.15PMN-PH-PT ceramics were grinded and polished to obtain flat and parallel surfaces for the following characterization. Volume density of the 0.15PMN-PH-PT ceramics was measured by Archimedes’ water immersion method. Rigaku D/max-2500/PC X-ray Diffractometer (XRD, Rigaku Corp., Japan) was used to characterize crystal structure. Silver electrode was formed on the ceramics surface by calcining the manually coated silver paste at 650 °C for 30 min for electrical properties measurement. Detailed dielectric, ferroelectric and piezoelectric performance measurement procedures were reported elsewhere [19–21].

3. Results and discussion

Figure 1 shows XRD patterns of the 0.15PMN-PH-PT ceramics with different compositions, in which the expanded XRD patterns around the [200] diffraction reflection and the deconvolution fitting of the [200] peak using the 0.15PMN-0.36PH-0.49PT ceramics as an example are also shown. As shown in figure 1(a), pure perovskite phase is obtained in all the synthesized 0.15PMN-PH-PT ceramics without detectable impurity within the detection sensitivity of the XRD measurement. From the expanded [200] diffraction reflection shown in figure 1(b), we can see that the diffraction peak changes gradually from symmetric and sharp singlet peak to doublet peaks accompanied by the variation of peaks’ intensity with decreasing the PH content. The [200] diffraction reflection of the 0.15PMN-0.36PH-0.49PT ceramics can be deconvolution fitted into rhombohedral (200)h peak, and tetragonal (002)T1 peak and (200)T2 peak, indicating the coexistence of rhombohedral phase and tetragonal phase (figure 1(c)). Such change regularity explains that the content of the tetragonal phase in the 0.15PMN-PH-PT system increases with decreasing the PH concentration, and all the sintered ceramics selected in the 0.15PMN-PH-PT system locate around the MPB composition.

Table 1 shows cell parameters determined based on the XRD data using the MDI Jade 6.5 software and density measured by the Archimedes method of the 0.15PMN-PH-PT ceramics. The 0.15PMN-PH-PT ceramics are dealt with as pseudo-cubic structure due to the coexistence of rhombohedral and tetragonal phases, whose content can not be distinguished accurately by the conventional XRD measurement. The cell volume increases first and then decreases with decreasing the PH content, whereas, the change regularity of bulk density and relative density is not obvious. However, the relative density of all the 0.15PMN-PH-PT ceramics exceeds 92%, especially the 0.15PMN-0.38PH-0.47PT ceramics, exhibiting the largest bulk density and relative density, being 8.53 g cm⁻³ and 97.76%, respectively. The high density provides effective guarantee for the 0.15PMN-PH-PT ceramics to obtain high resistivity, i.e., all exceeds 10¹² Ω·cm order, which can endure high electric-field poling to present excellent piezoelectricity (table 1).
The high densification can be observed by scanning electron microscopy (SEM) as shown in figure 2 using the 0.15PMN-0.4PH-0.45PT ceramics as an example. Due to using free surface of the ceramics after thermal etching at 825 °C for 30 min as SEM observation sample, slight depth of field and roughness, and residual PbZrO3 covering powder are observed. Basically, almost no closed pores, rather homogenous grains with clear grain boundary, and nearly round-shape grains, indicating partial densification effect induced by liquid-phase sintering mechanism \([22]\), are formed in the sintered ceramics, coinciding well with the high relative density. The grain size distributes rather centralized with average grain size of 3.4 μm basing on grain size statistics.

Figure 3 shows dielectric performance-temperature curves of the 0.15PMN-PH-PT ceramics measured at 1 kHz, which provide useful information on ferroelectric phase transition behavior of ferroelectric materials. All the 0.15PMN-PH-PT ceramics exhibit sole dielectric peak appearing at the Curie temperature \(T_m\), which relates to the pseudo-cubic MPB ferroelectric to cubic paraelectric phase transition depending on respective compositions. With the decrease of the PH content, the \(T_m\) temperature increases almost linearly, and the dielectric constant maximum \(\varepsilon_m\) presents extremum value at \(x = 0.40\), i.e., the 0.15PMN–0.40PH–0.45PT ceramics with 23208, corroborating the MPB effects in the 0.15PMN–PH–PT system \([23, 24]\).

Dielectric performance-temperature curves at different frequencies were measured to determine the ferroelectric characteristic of the 0.15PMN–PH–PT ceramics using the 0.15PMN–0.40PH–0.45PT ceramics as an
Table 1. Lattice cell parameters, densities and resistivity of the 0.15PMN-PH-PT ceramics.

| Composition          | a = b = c (Å) | α = β = γ (°) | Cell volume (Å³) | Bulk density (g cm⁻³) | Relative density (%) | Resistivity (Ω·cm) |
|----------------------|---------------|---------------|------------------|-----------------------|----------------------|-------------------|
| 0.15PMN-0.42PH-0.43PT| 4.07671       | 89.9027       | 67.75            | 8.25                  | 93.14                | 2.08 × 10¹²       |
| 0.15PMN-0.40PH-0.45PT| 4.09008       | 90.3175       | 68.42            | 8.51                  | 97.73                | 6.89 × 10¹²       |
| 0.15PMN-0.39PH-0.46PT| 4.08665       | 90.1590       | 68.25            | 8.46                  | 97.27                | 3.72 × 10¹²       |
| 0.15PMN-0.38PH-0.47PT| 4.07723       | 89.9348       | 67.78            | 8.53                  | 97.76                | 6.41 × 10¹²       |
| 0.15PMN-0.36PH-0.49PT| 4.05884       | 89.4591       | 66.86            | 8.13                  | 92.58                | 4.75 × 10¹²       |
The Curie-Weiss law fitting and quadratic law fitting using 1 kHz data above the T_m temperature range are shown in figure 5. As shown in figure 4, the T_m temperature maintains almost unchanged at different frequencies, i.e., from 291 °C at 1 kHz increasing to 292 °C at 2 MHz. The value of dielectric constant maximum (ε_m) decreases slightly around the T_m temperature, i.e., from 22598 to 19760 within the same frequency range, and the full width at half of ε_m is about 43 °C (257 °C–300 °C), indicating that the 0.15PMN-0.40PH-0.45PT ceramics may be normal ferroelectrics with some diffused phase transition characteristic [25, 26].

The calculated diffuse coefficient γ = 1.844, approaching 2, confirming their diffused phase transition characteristic but not typical relaxor ferroelectrics. Such diffused phase transition character is normally attributed to the existence of polar nano-regions (PNRs) (figure 5(b)) [25, 26].

Room-temperature polarization-electric field (P-E) hysteresis loops, and bipolar and unipolar strain-electric field (S-E) hysteresis curves of the 0.15PMN-PH-PT ceramics are shown in figures 6 and 7, respectively, and the corresponding ferroelectric and piezoelectric properties are displayed in table 2. All the 0.15PMN-PH-PT
ceramics show saturate P-E loops with slight asymmetry, which indicate that the ceramics are fully polarized, and the asymmetry can be attributed to the existence of point-defects induced space charge field due to the evaporation of some elements during sintering (figure 6) [12]. The full polarization can be confirmed further by the impedance spectrum measurement, where the phase angle approaches 90° [27] accompanied by large electromechanical coupling coefficient $K_p$ for all the 0.15PMN-PH-PT ceramics. The 0.15PMN-0.40PH-0.45PT ceramics present the best ferroelectricity, where the remnant polarization $P_r$ is 33.17 $\mu$C cm$^{-2}$ and coercive electric field $E_c$ is 7.62 kV cm$^{-1}$. Symmetrically butterfly-shaped S-E loops can be observed from figure 7(a), which relate to the motion and rotation of ferroelectric domains and domain wall boundaries, and show the excellent piezoelectricity of the 0.15PMN-PH-PT ceramics [20, 21]. Based on the unipolar S-E loops shown in figure 7(b), the converse piezoelectric constant $d_{33}^*$ and strain hysteresis can be calculated, being $d_{33}^* = S_{\text{max}} / E_{\text{max}}$ and strain hysteresis $= H_{E_{\text{max}} / S_{\text{max}}} \times 100\%$, respectively [28, 29]. With the increase of the PH content, the values of $S_{\text{max}}$ and $d_{33}^*$ increase first and then decrease, whereas strain hysteresis exhibits opposite change trend, i.e., decreases first and then increases. The 0.15PMN-0.38PH-0.47PT ceramics present the largest $d_{33}^*$ value and the least strain hysteresis value, also correlating with the MPB effects [23, 24].

Table 2 shows piezoelectricity of the 0.15PMN-PH-PT ceramics poled under 20 kV cm$^{-1}$. Piezoelectric constant $d_{33}$ and converse piezoelectric constant $d_{33}^*$ present similar change trend. With the increase of the PH content, the $d_{33}$ and $d_{33}^*$ values increase first and then decrease, corroborating the MPB effects. It is worth mentioning that the $d_{33}^*$ value is normally larger than the $d_{33}$ value [30, 31], whereas, in the 0.15PMN-PH-PT

![Figure 4](image-url) Figure 4. Temperature dependence of dielectric properties of the 0.15PMN-0.38PH-0.47PT ceramics measured at different frequencies upon heating.

![Figure 5](image-url) Figure 5. Linear fitting of the dielectric behavior of the 0.15PMN-0.38PH-0.47PT ceramics using the 1 kHz data according to (a) the Curie-Weiss law and (b) the quadratic law.
system, the $d_{33}^*$ value is far larger than the $d_{33}$ value, indicating that small external electric field can drive large induced strain. Furthermore, the S-E curves present almost linearly single-valued function behavior and far less than saturate, the strain hysteresis is relatively small, especially the $T_m$ temperature is rather high, therefore, the 0.15PMN-PH-PT ceramics present excellent prospect applications in high-temperature actuator fields. The electromechanical coupling coefficient $K_p$ may be affected more by density, which presents similar change trend to that of the relative density. Among which the 0.15PMN-0.38PH-0.47 ceramics have the best piezoelectric performance, where $d_{33} = 367.4 \text{ pC N}^{-1}, d_{33}^* = 716.7 \text{ pm V}^{-1}$ and $K_p = 53.42\%$.

Piezoelectric depolarization is a serious dilemma requiring special consideration for high-temperature piezoelectric applications. Figure 8 shows piezoelectricity thermal stability of the 0.15PMN-PH-PT ceramics poled under 20 kV cm$^{-1}$, from which the variation of piezoelectric constant with environmental temperature can be revealed clearly. All the 0.15PMN-PH-PT ceramics exhibit excellent temperature stability, and the piezoelectric performance decays little between 25 $^\circ$C–200 $^\circ$C temperature range, i.e., the values of $d_{33}$ and $K_p$ decrease rather less than 20%. These values decrease sharply at the ferroelectric to paraelectric phase transition temperature around 300 $^\circ$C, furthermore, the $T_C$ temperature increases gradually with decreasing the PH content. Therefore, the 0.15PMN-PH-PT ceramics possess excellent piezoelectric temperature stability and high depolarization temperature, which can be expected to be used in high-temperature piezoelectric actuator fields [8].
Table 2. Ferroelectric and piezoelectric properties of the 0.15PMN-PH-PT ceramics.

| Composition              | \( P_t (\mu\text{C cm}^{-2}) \) | \( E_c (\text{kV cm}^{-1}) \) | \( d_{33}^- (\text{pm V}^{-1}(15 \text{kV cm}^{-1}) \) | Strain Hysteresis (%) | \( d_{33}^+ (\text{pC N}^{-1})(20 \text{kV cm}^{-1}) \) | \( K_p (\%) (20 \text{kV cm}^{-1}) \) |
|-------------------------|---------------------------------|-------------------------------|-----------------------------------------------|----------------------|-----------------------------------------------|---------------------|
| 0.15PMN-0.36PH-0.49PT  | 10.79                           | 7.78                          | 613.3                                         | 26.29                | 282.6                                         | 45.87               |
| 0.15PMN-0.38PH-0.47PT  | 26.59                           | 7.20                          | 716.7                                         | 24.55                | 367.4                                         | 53.42               |
| 0.15PMN-0.39PH-0.46PT  | 25.40                           | 7.45                          | 683.3                                         | 28.92                | 350.8                                         | 52.75               |
| 0.15PMN-0.40PH-0.45PT  | 33.14                           | 7.62                          | 610.7                                         | 28.34                | 326.1                                         | 59.85               |
| 0.15PMN-0.42PH-0.43PT  | 27.29                           | 7.09                          | 522.2                                         | 37.62                | 203.2                                         | 45.50               |
4. Conclusions

High Tc temperature 0.15PMN-PH-PT piezoelectric ceramics were prepared by the conventional ceramics processing via the columbite precursor method. XRD analysis confirms that all the 0.15PMN-PH-PT ceramics synthesized locate around the MPB compositions, where rhombohedral and tetragonal ferroelectric phases coexist, and the tetragonal phase content increases with the decrease of the PH content. The 0.15PMN-0.38PH-0.47PT ceramics present the highest relative density of 97.76%. The Tm temperature increases almost linearly, and the $\varepsilon_m$ value increases first and then decreases with the decrease of the PH content in the 0.15PMN-PH-PT system ceramics, where the 0.15PMN-0.40PH-0.45PT ceramics present the largest $\varepsilon_m$ value of 25208 accompanied by $T_m = 278^\circ C$. The fitting Curie-Weiss constant C is of 10$^5$ magnitude, the dielectric peaks are slight broadening at different frequencies, and the quadratic law fitting diffuse coefficient $\gamma$ is larger than 1.8 in all the 0.15PMN-PH-PT ceramics, indicating that they are normal ferroelectrics with some diffused phase transition characteristic. The 0.15PMN-0.38PH-0.47PT ceramics exhibit the best electrical properties and excellent thermal stability, where $\varepsilon_m = 22598$, $T_m = 291^\circ C$, $P_r = 26.59 \mu C cm^{-2}$, $E_c = 7.20$ kV cm$^{-1}$, $d_{33} = 367.4$ pC N$^{-1}$, $d_{33}^* = 716.7$ pm V$^{-1}$, and $K_p = 53.42\%$, correlating with MPB effects and high densification. The 0.15PMN-PH-PT ceramics present good thermal stability, high depolarization temperature, and large low electric field induced strain and small hysteresis, providing excellent prospect applications in high-temperature actuator fields.

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