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Effects of CeO_2 addition on the piezoelectric properties of PNW–PMN–PZT ceramics

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Abstract

Temperature stability and electrical properties of the piezoelectric material are very important in piezoelectric transformers applications. In this study, it was investigated the temperature stability of PNW–PMN–PZT with CeO_2 additives and the variation of Zr/Ti ratio. Meanwhile, effects of CeO_2 additives and the variation of Zr/Ti ratio on the microstructure and electrical properties of PNW–PMN–PZT were investigated in detail. The results revealed that the optimized temperature stability of $\Delta f_r/f_{r25\,^\circ C} = 0.15\%$, $\Delta K_p/K_{p25\,^\circ C} = -0.86\%$ and $\Delta Q_m/Q_{m25\,^\circ C} = -45.26\%$ could be attained at $x = 0.1$ wt.% and Zr/Ti = 51/49. Moreover, optimized electrical properties were achieved: $K_p = 0.60$, $Q_m = 1405$, $d_{33} = 388$ pC/N, $\varepsilon_r = 2140$ and tan $\delta = 0.0059$. The obtained temperature stabilities and electrical properties make this composition a good candidate for high power piezoelectric transformer applications.

Keywords: B. X-ray methods; C. Electrical properties; C. Thermal properties; D. PZT

1. Introduction

Recently, piezoelectric transformers have been widely developed applying to the liquid display (LCD) backlight inverter, DC–DC converter and AC–DC converter, etc., because piezoelectric transformers have favorable characteristics such as miniaturized, low profile, high efficient power inverter and no electromagnetic noise compared with traditional electromagnetic transformers [1–3]. Since the piezoelectric transformers do not overheat during the using process, the piezoelectric ceramics should have a high factor Q_m and a low dielectric loss tan δ. Furthermore, the piezoelectric transformers were required to have good transform efficiency, which means its electromechanical coupling factor K_p must be high [4]. Finally d_{33} is also the figure of merit of the piezoelectric activity. Therefore piezoelectric material should have high piezoelectric constant (d_{33}). Besides, in order that piezoelectric materials can be applied to high power electronic apparatus, the temperature stability of piezoelectric properties of high power electronic apparatus must be required. Furthermore, for the ceramics used for piezoelectric transformers, the temperature stabilities of the resonant frequency ($\Delta f_r/f_{r25\,^\circ C}$) and electromechanical coupling factor ($\Delta K_p/K_{p25\,^\circ C}$) is further required to be low.

To achieve high electrical proprieties, many quaternary systems have been developed on the base of binary PZT system, such as PMN–PNN–PZT [5], PNW–PMS–PZT [6], etc. Otherwise, in order to improve electrical properties, the variations of Zr/Ti ratios and several dopants of either donor or acceptor type added separately together to PZT systems, such as CeO_2 [7], MnO_2 [8] and so on, have been intensively investigated. However, for the applications in high power piezoelectric transformers, only the high electrical properties is not enough and the better temperature stabilities were also requested. But there are few reports which simultaneously give the research on the electrical properties and temperature stabilities of PZT quaternary system.

In this work, the effects of CeO_2 doping and the variation of Zr/Ti ratios on the temperature stabilities of f_r, K_p and Q_m and the electrical properties of PNW–PMN–PZT ceramics were studied. The mechanisms for the changes of the electrical properties with CeO_2 doped and the variation of Zr/Ti ratios were also discussed. It is expected that the work can achieve the

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promising composition for high power piezoelectric transformers applications.

2. Experimental procedures

The powders and ceramics with compositions of $0.90Pb(Zr_yTi_{1-y})O_3$–$0.07Pb(Mn_{1/3}Nb_{2/3})O_3$–$0.03Pb(Ni_{1/2}W_{1/2})O_3 + x$ wt.%CeO_2 (PNW–PMN–PZT) were prepared via a conventional mixed-oxide process, where $x = 0.0$, 0.1, 0.2, 0.3 and 0.4 and $y = 52/48$, 51/49, 50/50, 49/51, 48/52, respectively. Reagent-grade oxide powders Pb_3O_4 (97%), ZrO_2 (99%), TiO_2 (98%), Nb_2O_5 (99.5%), CeO_2 (99.5%), NiO (99%), WO_3 (99.9%), MnO_2 (99%) were mixed, respectively. The mixtures were ball-milled in ethanol for 12 h, and then dried at 80 °C. The powders were calcined at 800 °C for 2 h. After calcining, the powders were added with 5 wt.% polyvinyl alcohol (PVA) solution, and then were pressed into pellets with 15 mm in diameter at 100 MPa. Pellets were sintered at 1150 °C for 4 h in a sealed alumina crucible with lead atmosphere. Subsequently, the sintered discs were polished and silver-paste electrodes were fired at 850 °C, respectively. The ceramics were poled in the silicone oil at 120 °C by applying a DC electric field of 3 kV/mm for 30 min.

The microstructure of surface of as-sintered ceramics was observed with a scanning electron microscopy (SEM, Model Quanta 200, FEI Company). The bulk density was measured with the Archimedes method. Dielectric properties were obtained by measuring the capacitance and dielectric loss at 1 kHz at room temperature with an LCR meter (Model HP4294A). The piezoelectric constants (d_{33}) were measured with a quasi-static piezoelectric d_{33} meter (Model ZJ-3d, Institute of Acoustics Academic Sinica, China). The electromechanical coupling factor (K_p) and mechanical quality factor (Q_m) were calculated by the resonance and anti-resonance technique on the basis of IEEE standards using an impedance analyzer (Model HP4294A). The temperature coefficients of the piezoelectric properties were measured in the temperature range from −20 to +80 °C. The equation used for $\Delta f_r/f_{r25\,^{\circ}C}$, $\Delta K_p/K_{p25\,^{\circ}C}$ and $\Delta Q_m/Q_{m25\,^{\circ}C}$ calculations are as follows:

$$\frac{\Delta f_r}{f_{r\,25\,^{\circ}C}}(\%) = \frac{f_r - f_r(25\,^{\circ}C)}{f_r(25\,^{\circ}C)} \times 100 \qquad (1)$$

$$\frac{\Delta K_p}{K_{p\,25\,^{\circ}C}}(\%) = \frac{K_p - K_p(25\,^{\circ}C)}{K_p(25\,^{\circ}C)} \times 100 \qquad (2)$$

$$\frac{\Delta Q_m}{Q_{m\,25\,^{\circ}C}}(\%) = \frac{Q_m - Q_m(25\,^{\circ}C)}{Q_m(25\,^{\circ}C)} \times 100 \qquad (3)$$

3. Results and discussion

3.1. Microstructure and density

Fig. 1 shows the SEM surface photographs of CeO_2-doped PNW–PMN–PZT ceramics. It can clearly be seen that the grain size of undoped PNW–PMN–PZT ceramics was about 3.1 μm. The specimen doped with 0.1 wt.% CeO_2 exhibited the largest grain size about 4.8 μm and more uniform structure. However, further increasing amount of CeO_2 above 0.2 wt.% reduced the grain size gradually. From the above results, it is concluded that

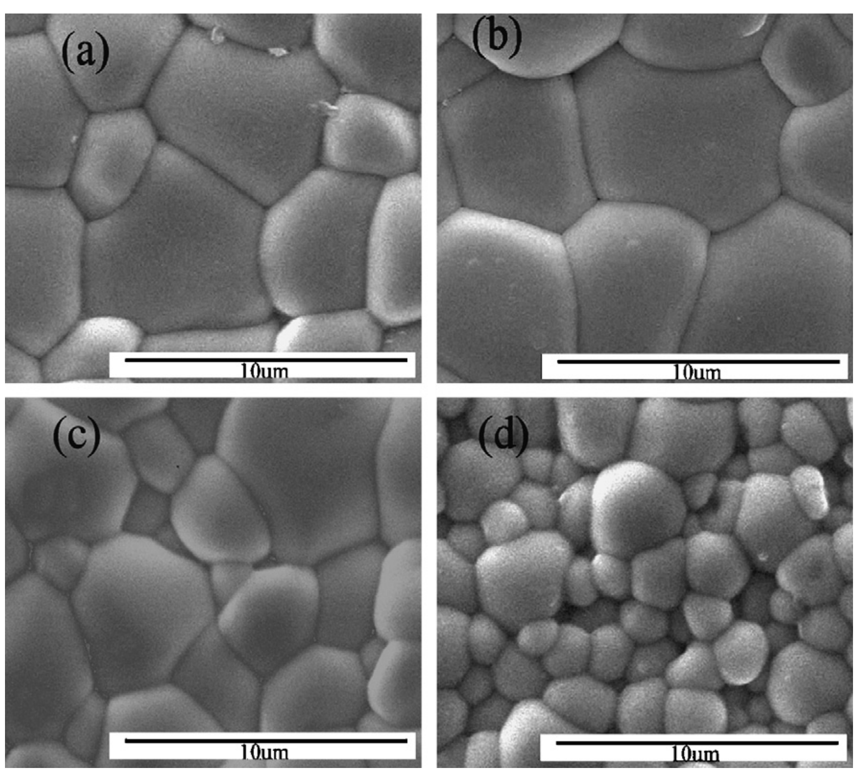

Fig. 1. The SEM photographs of PNW–PMN–PZT ceramics with CeO_2 additives: (a) 0.0 wt.%; (b) 0.1 wt.%; (c) 0.2 wt.%; (d) 0.4 wt.%.

CeO_2 has solubility of about 0.1 wt.% in PNW–PMN–PZT system. It is may be that Ce ion is homogenously dissolved in PNW–PMN–PZT system when the CeO_2 addition is below 0.1 wt.%, but further addition of CeO_2 will inhibit the grain growth due to accumulation of Ce ion at the grain boundary.

Fig. 2 shows the bulk density of ceramics as a function of CeO_2 content. With increasing CeO_2 content, the density increases at first and reaches the maximum value (7.87 g/cm^3) at $x = 0.1$ wt.%, and then decreases. When the content of CeO_2 is in the range of 0.2–0.3 wt.%, the shift of the bulk density is slight. The result indicates that the proper addition of CeO_2 can enhance the density of ceramics. According to the report of Hou et al. [9] that the variation of SEM is in connect with the result of density. When the content of CeO_2 is at 0.1 wt.%, well-grown grains with relatively homogenous size can be seen from the SEM photograph of the ceramics. This homogenous microstructure may be beneficial to the improvement of density and results in the highest density at 0.1 wt.%.

3.2. Piezoelectric and dielectric properties

3.2.1. PNW–PMN–PZT + x wt.%CeO_2

Fig. 3 shows the changes in K_p, Q_m and d_{33} of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 as a function of CeO_2 content. When the content of CeO_2 is below 0.1 wt.%, K_p and d_{33} increases, and Q_m decreases. The variations may be due to CeO_2 is a dopant with the characteristics of both 'soft' and 'hard' and Ce^{4+} is reduced during calcinations or sintering from the Ce^{4+} to the Ce^{3+} state [7]. When the Ce^{3+} occupies the A-site of perovskite structure, it leads to lead vacancies that make the mobility of the domain wall easier. Subsequently, K_p and d_{33} increases, but Q_m decreases. When the content of CeO_2 is in the range of 0.1–0.2 wt.%, K_p and d_{33} decreases, and Q_m increases, which attributes that Ce^{3+} entered into the B site as an acceptor dopant. As well known, the substitutions of B site by acceptor dopant would lead to the creation of oxygen vacancies, which inhibit the movement of domain wall, and result in the decrease of K_p and d_{33}, and Q_m increases. As the content of CeO_2 is above 0.2 wt.%, K_p, d_{33} and Q_m gradually decreases. The decrease of K_p and d_{33} may be caused by the decrease in the grain size [10]. While excess CeO_2 will stay in the boundary and form a grain boundary layer, which cause the decrease of Q_m [11,12]. Considering the above variations of the three parameters for the request of the applications with increasing CeO_2 content, the best values of K_p (0.55), Q_m (1722) and d_{33} (342 pC/N) are simultaneously obtained at $x = 0.1$ wt.%.

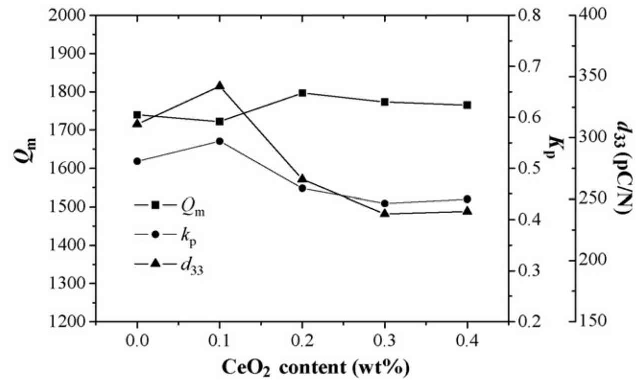

Fig. 3. K_p, Q_m and d_{33} of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives.

Fig. 4 shows the changes in ε_r and tan δ of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 as a function of CeO_2 content. When the content CeO_2 is below 0.1 wt.%, ε_r and tan δ increase with increasing the CeO_2 content. It is attributed to the creation of lead vacancies, which makes the movement of the ferroelectric domain walls easily and results in the increase of ε_r and tan δ. When the content of CeO_2 is in the range of 0.1–0.2 wt.%, ε_r and tan δ increase gradually with the content of CeO_2 increasing. It is caused by the creation of oxygen vacancies, which pins the movement of the ferroelectric domain walls and results in the increase of ε_r and tanδ. When the content CeO_2 is more than 0.2 wt.%, the excess CeO_2 will stay in the boundary and form a grain boundary layer, ε_r decreases and tan δ increases because of grain boundary layer. Considering the above variations of the two parameters for the request of the applications with increasing CeO_2 content, the best ε_r value (1933) and tan δ value (0.0057) are obtained at $x = 0.1$ wt.%.

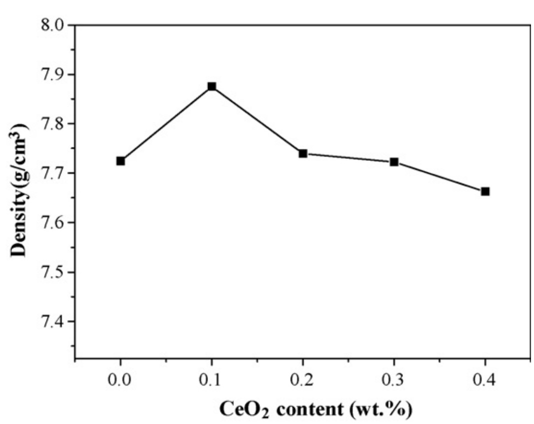

Fig. 2. Density of PNW–PMN–PZT ceramics with CeO_2 additives.

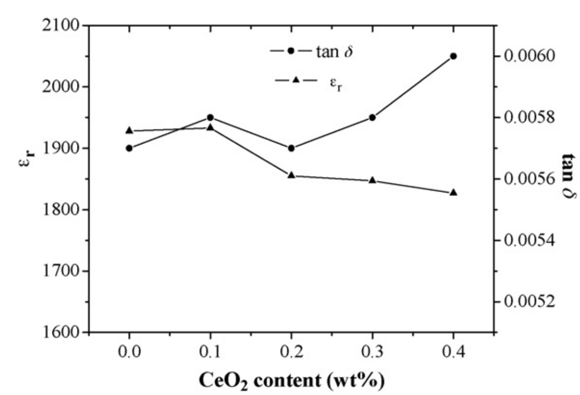

Fig. 4. ε_r and tan δ of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives.

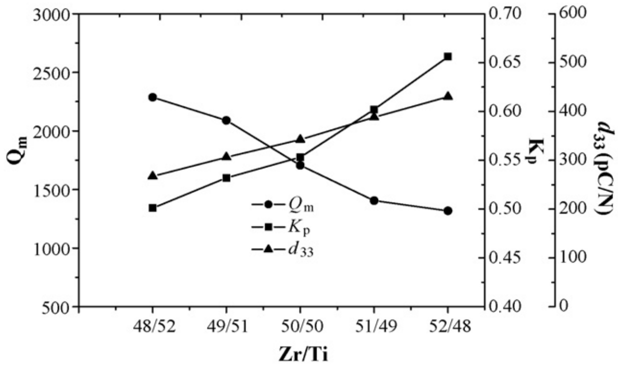

Fig. 5. K_p, Q_m and d_{33} of the ceramics with 0.1 wt.%CeO_2 as a function of different Zr/Ti ratios.

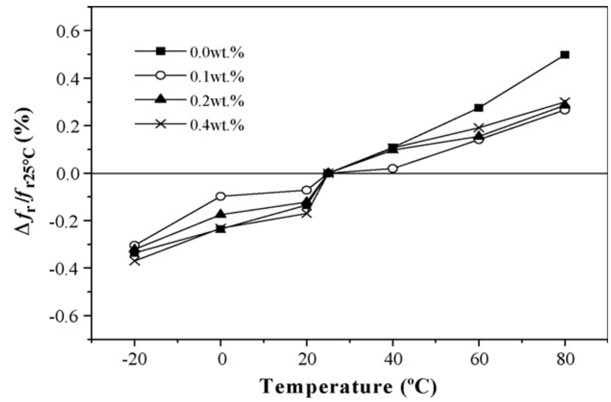

Fig. 7. Temperature dependence of f_r of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives.

3.2.2. PNW–PMN–PZT + 0.1 wt.%CeO_2 ceramics of different Zr/Ti ratios

Figs. 5 and 6 show the changes in K_p, Q_m, d_{33}, ε_r and tan δ of the ceramics with 0.1 wt.%CeO_2 additive as a function of Zr/Ti ratios. As can be seen from Figs. 5 and 6, we can find that the K_p, d_{33}, ε_r and tan δ increase and the Q_m decreases with increasing the Zr/Ti ratios. The maximum values of K_p, d_{33} and ε_r and the minimum value of Q_m are obtained at Zr/Ti = 52/48. The result is related to its phase situated nearby MPB, at the MPB, the materials can be both tetragonal and rhombohedral phase, and the spontaneous polarization can be oriented along any 1 of these 14 directions and it is easy for domains motion in poling and resulted in high piezoelectric and dielectric properties [13]. Therefore, its values of K_p, d_{33} and ε_r show the maximum. In contrast, unlike K_p, d_{33} and ε_r, Q_m exhibited a minimum value. This is understandable since tetragonal phases and rhombohedral phase coexisted near the MPB and thus can move easily.

Taken into consideration of the material in high power piezoelectric transformers applications, we wish the material that can attain the high K_p, Q_m, d_{33} and low tan δ. Therefore we chose that the optimized electrical properties of the ceramics with 0.1 wt.%CeO_2 and Zr/Ti = 51/49, which were as follows: $K_p = 0.60$, $Q_m = 1405$, $d_{33} = 388$ pC/N, $\varepsilon_r = 2140$ and tan $\delta = 0.0059$.

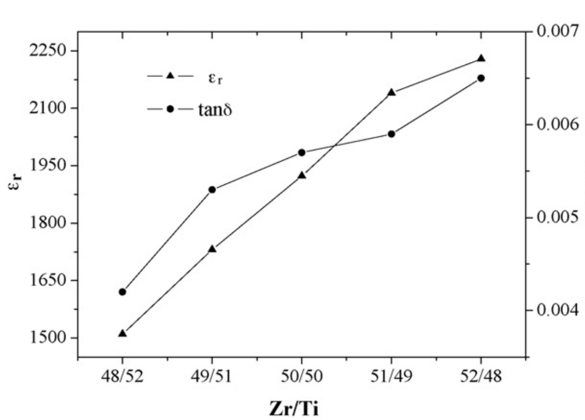

Fig. 6. ε_r and tan δ of the ceramics with 0.1 wt.%CeO_2 as a function of different Zr/Ti ratios.

3.3. Temperature stability

3.3.1. PNW–PMN–PZT + x wt.%CeO_2

For practical applications in high power piezoelectric transformers, it is essential for the $\Delta f_r/f_{r25\,^\circ C}$ and $\Delta K_p/K_{p25\,^\circ C}$ to change slightly under fluctuating thermal environment. So the $\Delta f_r/f_{r25\,^\circ C}$ and $\Delta K_p/K_{p25\,^\circ C}$ are two important performance parameters that should be taken into consideration and studied. Fig. 7 shows temperature dependence of f_r of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives. The temperature coefficient was calculated by resonant frequency of the planar vibration mode measured in the range of -20 to 80 °C. From Fig. 7, at the same temperature, the $\Delta f_r/f_{r25\,^\circ C}$ decreases for the addition of 0.0–0.1 wt.% CeO_2, then increases with the CeO_2 content above 0.1 wt.%. The minimum value (0.5%) of $\Delta f_r/f_{r25\,^\circ C}$ is obtained at $x = 0.1$ wt.%.

Fig. 8 shows temperature dependence of K_p of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives, similarly, in the temperature from -20 to 80 °C. From Fig. 8, at the same temperature, the $\Delta K_p/K_{p25\,^\circ C}$ decreases for the addition of 0.0–0.1 wt.% CeO_2 and increases for the addition of 0.1–0.4 wt.% CeO_2. The minimum of $\Delta K_p/K_{p25\,^\circ C}$ was obtained at the addition of 0.1 wt.% CeO_2 and the value is -1.80%.

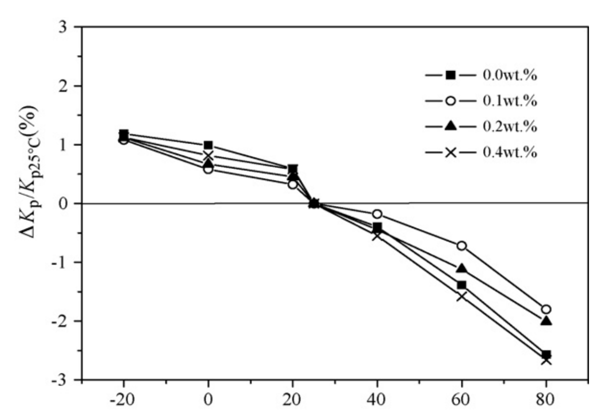

Fig. 8. Temperature dependence of K_p of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives.

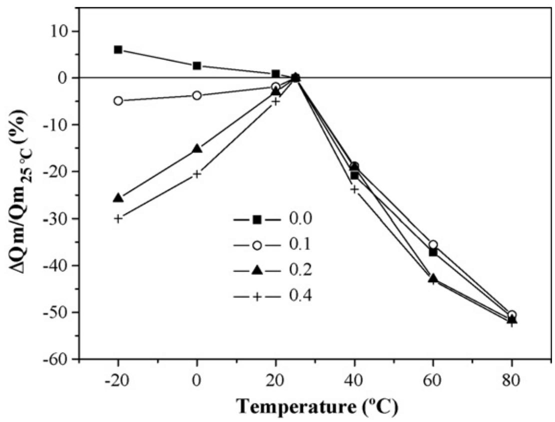

Fig. 9. Temperature dependence of Q_m of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives.

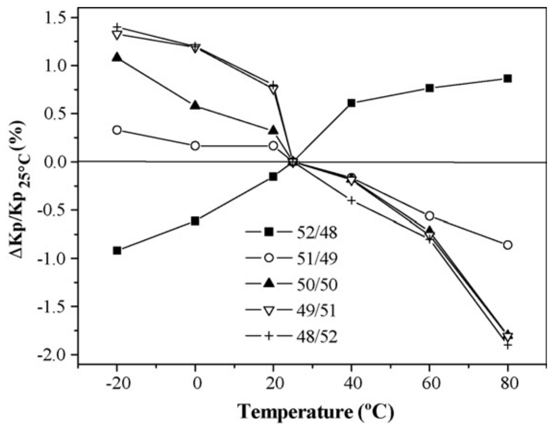

Fig. 11. Temperature dependence of K_p of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios.

Fig. 9 shows temperature dependence of Q_m of PNW–PMN–PZT ceramics at Zr/Ti = 50/50 with CeO_2 additives. From Fig. 9, it can be seen that the $\Delta Q_m/Q_{m25\,^\circ C}$ of the undoped samples increased with the increasing of temperature. The $\Delta Q_m/Q_{m25\,^\circ C}$ of the doped samples decreased at first at the range from −20 to 25 °C. And then the $\Delta Q_m/Q_{m25\,^\circ C}$ of the doped sample increased quickly at the range from 25 to 80 °C. The minimum of $\Delta Q_m/Q_{m25\,^\circ C}$ was obtained at the addition of 0.1 wt.% CeO_2 and the value is −50.25%.

3.3.2. PNW–PMN–PZT + 0.1 wt.%CeO_2 ceramics of different Zr/Ti ratios

Fig. 10 shows temperature dependence of f_r of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios. From Fig. 9, in the temperature range from −20 to 80 °C, it can be seen that the $\Delta f_r/f_{r25\,^\circ C}$ shifted from plus (+) into minus (−) value with increasing Zr/Ti ratios. When the Zr/Ti ratios at rich Zr side, the $\Delta f_r/f_{r25\,^\circ C}$ decreases with the Zr/Ti ratios increases at the range from −20 to 80 °C. When the Zr/Ti ratios at Zr poor side, the $\Delta f_r/f_{r25\,^\circ C}$ increases with the Zr/Ti ratios increases at the range from −20 to 80 °C. The minimum change ($\Delta f_r/f_{r25\,^\circ C}$ = 0.15%) is attained at Zr/Ti = 51/49.

Fig. 11 shows temperature dependence of K_p of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios. From Fig. 10, in the temperature range from −20 to 80 °C, it can be seen that the $\Delta K_p/K_{p25\,^\circ C}$ from minus (−) into plus (+) value with increasing Zr/Ti ratios. The minimum change ($\Delta K_p/K_{p25\,^\circ C}$ = −0.86%) is obtained at in the vicinity of the MPB (Zr/Ti = 51/49).

Fig. 12 shows temperature dependence of Q_m of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios. From Fig. 12, it can be seen that the $\Delta Q_m/Q_{m25\,^\circ C}$ of Zr/Ti = 51/49–52/48 increased at the range from −20 to 80 °C. The $\Delta Q_m/Q_{m25\,^\circ C}$ of Zr/Ti = 48/52–50/50 decreased slowly at the range from −20 to 25 °C. When the temperature is at the range from 25 to 80 °C, the $\Delta Q_m/Q_{m25\,^\circ C}$ of Zr/Ti = 48/52–50/50 increased quickly. The minimum value ($\Delta Q_m/Q_{m25\,^\circ C}$ = −45.26%) was obtained at Zr/Ti = 51/49.

It is clear that $\Delta f_r/f_{r25\,^\circ C}$, $\Delta K_p/K_{p25\,^\circ C}$ and $\Delta Q_m/Q_{m25\,^\circ C}$ are more slightly with the addition of 0.1 wt.% CeO_2 and Zr/Ti = 51/49 than those of others, which suggests piezoelectric ceramics with 0.1 wt.% CeO_2 addition and Zr/Ti = 51/49 have the optimized stability for high power piezoelectric transformer applications. At the same time, the optimized electrical properties are obtained, which makes the piezoelectric ceramics at x = 0.1 wt.% and Zr/Ti = 51/49 to be

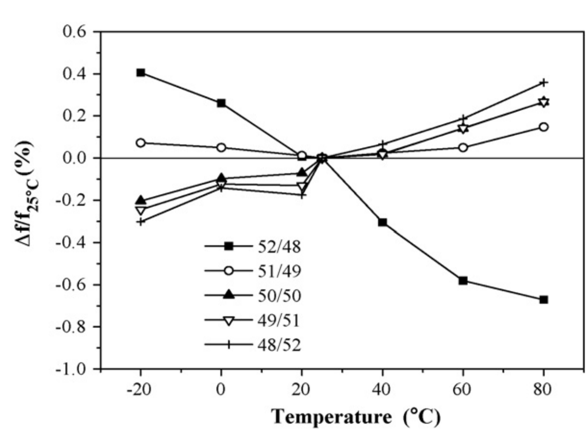

Fig. 10. Temperature dependence of f_r of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios.

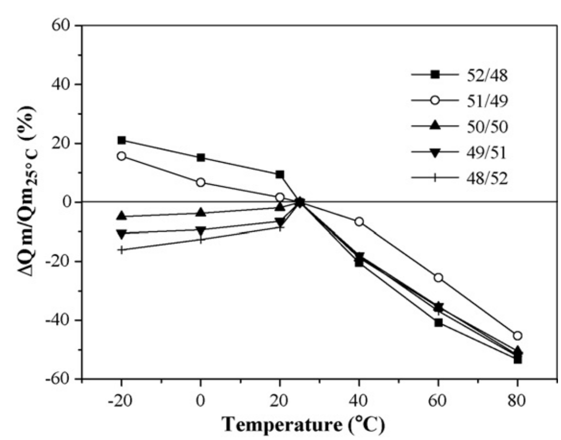

Fig. 12. Temperature dependence of Q_m of the ceramics with 0.1 wt.%CeO_2 for different Zr/Ti ratios.

relatively suitable for high power piezoelectric transformers applications.

4. Conclusions

The PNW–PMN–PZT piezoelectric ceramics with CeO_2 addition were synthesized by the conventional mixed-oxide method. The effects of CeO_2 addition and the variation of Zr/Ti ratios on piezoelectric, dielectric properties and temperature stabilities of PNW–PMN–PZT quaternary piezoelectric ceramics were discussed. The results are summarized as follows:

(1) The largest density and the uniformer grain size was achieved at $x = 0.1$ wt.%. Further increasing CeO_2 content to 0.4 wt.% caused the density and the grain size to decrease.
(2) PNW–PMN–PZT with 0.1 wt.% CeO_2 addition and Zr/Ti = 51/49 exhibited favorable piezoelectric and dielectric properties, which are $K_p = 0.60$, $Q_m = 1405$, $d_{33} = 388$ pC/N, $\varepsilon_r = 2140$ and tan $\delta = 0.0059$.
(3) The temperature stability of $\Delta f_r/f_{r25\,^\circ C}$, $\Delta K_p/K_{p25\,^\circ C}$ and $\Delta Q_m/Q_{m25\,^\circ C}$ of the ceramics at $x = 0.1$ wt.% and Zr/Ti = 51/49 show the lowest values among all the samples, which are $\Delta f_r/f_{r25\,^\circ C} = 0.15\%$, $\Delta K_p/K_{p25\,^\circ C} = -0.86\%$ and $\Delta Q_m/Q_{m25\,^\circ C} = -45.26\%$.

Taken into consideration the temperature stability and the electrical properties, the results show that the PNW–PMN–PZT + 0.1 wt.% CeO_2 and Zr/Ti = 51/49 of piezoelectric material are good candidate for high power piezoelectric transformer applications.

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