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## Enhanced mechanical quality factor of $\text{BiScO}_3$ – $\text{PbTiO}_3$ piezoelectric ceramics using glass composition

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Compared with pure Pb-based perovskite ferroelectric materials,  $\text{Bi}(\text{Me})\text{O}_3$ – $\text{PbTiO}_3$  ( $\text{Me} = \text{Sc}^{3+}, \text{In}^{3+}, \text{Yb}^{3+}$ ) have attracted attention due to their remarkable advantage in their Curie temperature. Among them,  $\text{BiScO}_3$ – $\text{PbTiO}_3$  piezoelectric ceramic is a potential piezoelectric material in high-temperature applications for its high Curie temperature and excellent piezoelectric coefficient. However, its shortcomings are high dielectric loss and low mechanical quality factor. Herein, we report the improvement of the mechanical quality factor of 0.36BS–0.64PT ceramics through the addition of glass composition ( $\text{GeO}_2$ ). There is a small change in the Curie temperature after  $\text{GeO}_2$  addition. The piezoelectric coefficient  $d_{33}$  and planar electromechanical coupling factor  $k_p$  increase first and then decrease, and the mechanical quality factor  $Q_m$  monotone increases with an increase in  $\text{GeO}_2$ . The 0.36BS–0.64PT + 0.5 mol% $\text{GeO}_2$  ceramics have optimal electrical properties with  $T_C$  of 455 °C,  $d_{33}$  of 385 pC N<sup>-1</sup>,  $k_p$  of 58%, and  $Q_m$  of 90. In addition, the thermal stability of 0.36BS–0.64PT +  $x\text{GeO}_2$  and 0.36BS–0.64PT ceramics is almost the same. It was concluded that the mechanical quality factor of BS–PT ceramics can be enhanced by the addition of  $\text{GeO}_2$  with other properties remaining unchanged.

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### 1. Introduction

With the requirement of high-temperature piezoelectric sensors and actuators in aerospace, automobiles, oil and gas exploration fields, high-temperature piezoelectric materials have received unprecedented attention.<sup>1–4</sup> The use temperature of piezoelectric materials is about half of the Curie temperature ( $T_C$ ) due to piezoelectric performance degradation at high temperatures.<sup>5</sup> The commercial lead-zirconium-titanium (PZT) piezoelectric ceramics have been studied for decades and are widely used in piezoelectric sensors and transducers, but these ceramics cannot be used in environments above 200 °C, while applications in the aerospace and automotive industries require piezoelectric devices to operate at a temperatures of 300 °C or higher.<sup>6,7</sup> Therefore, the development of new high-temperature-stable piezoelectric materials is an extremely urgent need.

Bismuth-based perovskite solid solutions with the formula of  $\text{Bi}(\text{Me})\text{O}_3$ – $\text{PbTiO}_3$  ( $\text{Me} = \text{Sc}, \text{In}, \text{Yb}, \text{Mg/Ti}, \text{Fe}, \text{Ga}$  *et al.*), which are potential alternatives for high-temperature application, have attracted growing interest due to their ultra-high Curie temperature.<sup>8–13</sup> Among them,  $\text{BiYbO}_3$ – $\text{PbTiO}_3$ ,  $\text{BiInO}_3$ – $\text{PbTiO}_3$ ,  $\text{Bi}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$ – $\text{PbTiO}_3$  and  $\text{BiFeO}_3$ – $\text{PbTiO}_3$  are not suitable for

applications due to their poor piezoelectric properties.  $\text{BiScO}_3$ – $\text{PbTiO}_3$  (BS–PT) solid solutions have drawn considerable attention because of their high piezoelectric constant ( $d_{33}$ ) of 460 pC N<sup>-1</sup> and high  $T_C$  of 450 °C near the morphotropic phase boundary (MPB).<sup>14,15</sup> In addition, due to the advantages of high working efficiency, good flexibility, high strength and toughness of flexible ceramic film materials, some researchers have focused on preparing multifunctional materials on flexible substrates. For example, Liu successfully fabricated  $\text{Mn}-\text{In}_2\text{O}_3$  thin films with excellent mechanical durability and transparent conductive oxide  $\text{SrVO}_3$  thin films by pulsed laser deposition (PLD) on flexible substrates.<sup>16,17</sup> BS–PT solid solutions have small crystal anisotropy and excellent electrical performance, which can be combined with a flexible substrate to improve the piezoelectric response.<sup>18</sup>

The shortcomings of BS–PT ceramics are high dielectric loss ( $\tan \delta > 3\%$ ) and a low mechanical quality factor ( $Q_m < 30$ ),<sup>19,20</sup> which limit their application at high temperatures. Previously, a great deal of work has been done to increase the  $Q_m$  of BS–PT ceramics through chemical methods, such as the addition of  $\text{Mn}$ ,  $\text{Fe}$ ,  $\text{Pb}(\text{Mn}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ,  $\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3$ .<sup>4,21–25</sup> The mechanical quality factor can be enhanced by chemical methods, while the piezoelectric performance and Curie temperature are often decreased. For example, the piezoelectric performance of  $(0.97-x)\text{BiScO}_3$ – $x\text{PbTiO}_3$ – $0.03\text{Pb}(\text{Mn}_{1/3}\text{Nb}_{2/3})\text{O}_3$  ceramics deteriorated severely ( $d_{33} < 300$  pC N<sup>-1</sup>).<sup>24</sup> The Curie temperature of  $0.03\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3$ – $0.33\text{BS}$ – $0.64\text{PT}$  and

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$\text{BiScO}_3\text{-PbTiO}_3\text{-Pb}(\text{Yb}_{1/2}\text{Nb}_{1/2})\text{O}_3$  ceramics is less than 400 °C.<sup>23,26</sup> Mn and Fe co-modified BS-PT ceramics show both a lower  $d_{33}$  of 235 pC N<sup>-1</sup> and dielectric constant  $\epsilon_r$  of 815 compared with pure BS-PT ceramics.<sup>25</sup>

$\text{GeO}_2$ -glass former is an additive used in glass. This glass former can decrease the dielectric loss and increase insulativity.  $\text{Ge}^{4+}$  ions tend to segregate along the grain boundaries, leading to a reduction in the jumping of mobile ions, which in turn causes a decrease in the conductivity of the ceramics.<sup>27</sup> Mou *et al.* used  $\text{Ge}^{4+}$  to enhance the piezoelectric performance and adjusted dielectric properties.<sup>28</sup>  $\text{GeO}_2$  can synchronously enhance the  $Q_m$  and piezoelectric properties in the KNN and BCZT system.<sup>29,30</sup> Therefore, we investigated the effect of  $\text{GeO}_2$  addition on the phase structure, dielectric, piezoelectric, ferroelectric and electromechanical coupling properties of BS-PT ceramics.

## 2. Experimental procedures

The  $\text{GeO}_2$  doped 0.36BS-0.64PT piezoelectric ceramics (abbreviated as 0.36BS-0.64PT +  $x\text{GeO}_2$ ,  $x$  = 0 mol%, 0.25 mol%, 0.5 mol%, 0.75 mol%, 1.0 mol%, 1.5 mol%) were prepared by the conventional solid-state reaction method. Starting materials were  $\text{Bi}_2\text{O}_3$  (99.99%),  $\text{Sb}_2\text{O}_3$  (99.99%),  $\text{PbO}$  (99.9%),  $\text{TiO}_2$  (99.5%), and  $\text{GeO}_2$  (99.99%). These oxide powders were weighed and mixed according to the chemical formula, adding 1 mol%  $\text{Bi}_2\text{O}_3$  and  $\text{PbO}$  to compensate for evaporation during the sintering process. The mixture was ball milled for 12 h in absolute ethyl alcohol (99.7%) with zirconia balls, then dried and pressed into pellets at 60 MPa. The pellets were placed in a crucible and calcined at 780 °C for 5 h at a heating/cooling rate of 5 °C min<sup>-1</sup>. Thereafter, the  $\text{GeO}_2$  of different amounts was added to the calcined powder and ball milled again for another 24 h. After ball milling, the slurry was dried, crushed, and mixed with 5% polyvinyl alcohol binder (PVA). The powder was passed through a 200 mesh sieve and was pressed into pellets of 8 mm in diameter and 1.5 mm in thickness under an actual pressure of 200 MPa. Subsequently, it was heated at 600 °C for 2 h to eliminate the binder, and sintered in a sealed  $\text{Al}_2\text{O}_3$  crucible in the range of 1000–1100 °C for 2 h to form ceramic samples.

The crystal structure of sintered ceramics was analyzed on an X-ray diffractometer (Rigaku, Miniflex600, Japan) with  $\text{Cu-K}\alpha$

radiation. The TEM was performed using a field emission transmission electron microscope (TEM, FEI Tecnai F20, USA). A scanning electron microscope (SEM, SU8010, Hitachi, Japan) was used to investigate the microstructure. The density of the sintered samples was determined by the Archimedes method. The obtained ceramics were polished to a thickness of 0.5 mm, and then coated with silver paste on the surface to form the electrodes. The samples were poled in an oil bath at 120 °C in an electric field of 40 kV cm<sup>-1</sup> for 15 min. After standing for 24 h, the electrical properties were tested. The dielectric constant ( $\epsilon_r$ ) and dielectric loss ( $\tan \delta$ ) were measured on an impedance analyzer (E4990A, Agilent Technologies, USA), and the planar electromechanical coupling factor ( $k_p$ ) and  $Q_m$  were calculated according to the resonance and anti-resonance method. Ferroelectric hysteresis loop measurements were performed on an aix-ACCT TF2000 ferroelectric analyzer ( $f$  = 1 Hz). Piezoelectric coefficients were measured by a quasi-static  $d_{33}$  meter (Institute of Acoustics, Chinese Academy of Sciences, model ZJ-4AN).

## 3. Results and discussion

### 3.1 Structure and phase analysis

Fig. 1(a) shows the X-ray diffraction patterns (log scale) of powder 0.36BS-0.64PT +  $x\text{GeO}_2$  ceramics. The main peaks of all samples matched with the perovskite phase, indicating formed stable perovskite solid solutions. It can be easily seen that the second phase ( $\text{GeO}_2$ ) appeared as the doping content of  $\text{GeO}_2$  increased, indicating that it is difficult for  $\text{Ge}^{4+}$  to enter the lattice. The radius of  $\text{Ge}^{4+}$  ion (0.53 Å for CN = 6) is almost similar to that of  $\text{Ti}^{4+}$  ions (0.605 Å for CN = 6), and it is considered that  $\text{Ge}^{4+}$  ions can enter the B site of the perovskite unit cell and substitute  $\text{Ti}^{4+}$  ions.<sup>31</sup> Fig. 1(b) shows the amplified images of (200)<sub>c</sub> peaks. It can be easily seen that the phase structure is slightly changed after adding  $\text{GeO}_2$ . The lattice parameters of 0.36BS-0.64PT +  $x\text{GeO}_2$  ceramics were calculated according to the diffraction patterns, as shown in Fig. 1(c). With the increase in the  $\text{GeO}_2$  content, the “ $c$ ” value decreases continuously while the “ $a$ ” value increases, and the  $c/a$  value keeps decreasing from 1.023 to 1.014, which confirms that the  $\text{Ge}^{4+}$  ion entered the B site of the perovskite unit cell.

Fig. 2 shows the TEM image of 0.36BS-0.64PT + 0.5GeO<sub>2</sub> ceramics. One set of lattice fringes with an interplanar spacing

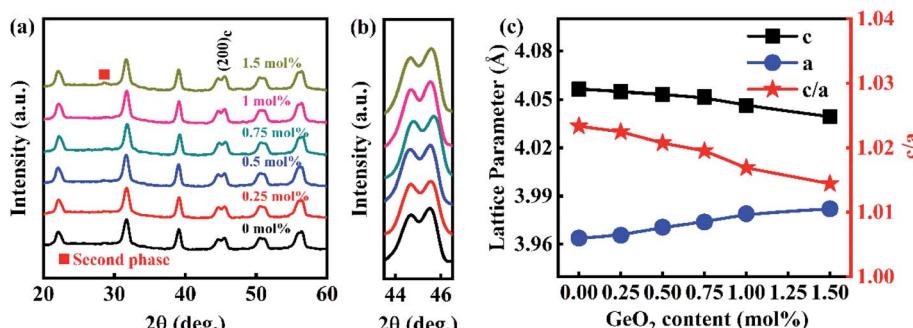


Fig. 1 (a) X-ray diffraction patterns (log scale) of powder 0.36BS-0.64PT +  $x\text{GeO}_2$  ceramics. (b) The amplified images of (200)<sub>c</sub> peaks. (c) Lattice parameters of 0.36BS-0.64PT +  $x\text{GeO}_2$  ceramics.



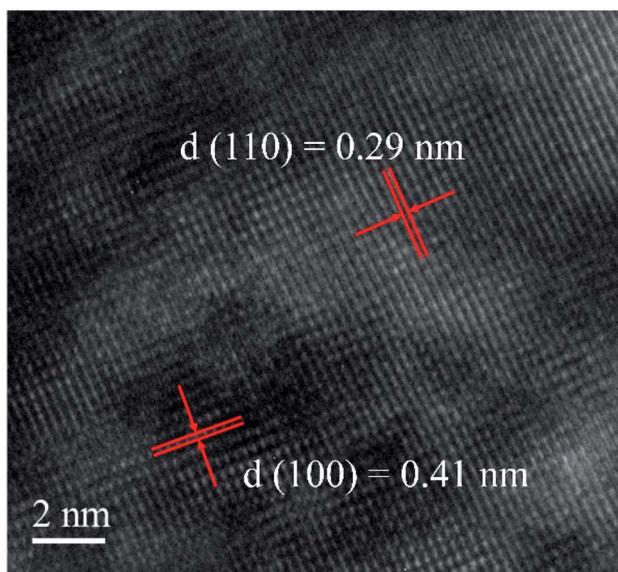


Fig. 2 TEM image of 0.36BS–0.64PT + 0.5GeO<sub>2</sub> ceramics.

of 0.29 nm, corresponding to the (110) plane of the perovskite phase, and another set of lattice fringes with a 0.41 nm interplanar spacing, corresponding to the (100) plane of the perovskite phase, which is consistent with the phase structure shown by the XRD results.

Fig. 3 shows the microstructure of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics sintered at 1080 °C for 2 h. These images clearly show the morphology of the ceramic samples with few holes, dense grains and clear grain boundaries. Table 1 lists the density of the 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics. The actual density ( $\rho_1$ ) was calculated by the Archimedes method, and the theoretical density ( $\rho_2$ ) was calculated according to XRD data. The relative density ( $\rho_r$ ) is 96.17–97.82%, showing a high density. The insets of Fig. 3 show the statistical data of the grain size of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics. With the increase in GeO<sub>2</sub>, the average grain size decreases from 8.46  $\mu$ m to 2.54  $\mu$ m, indicating that GeO<sub>2</sub> can inhibit the growth of grains. A similar phenomenon has been reported in other Ge doped ceramics. Fig. 3(e and f) show that a small amount of GeO<sub>2</sub> gathers at the grain boundary, which hinders the movement of the grain

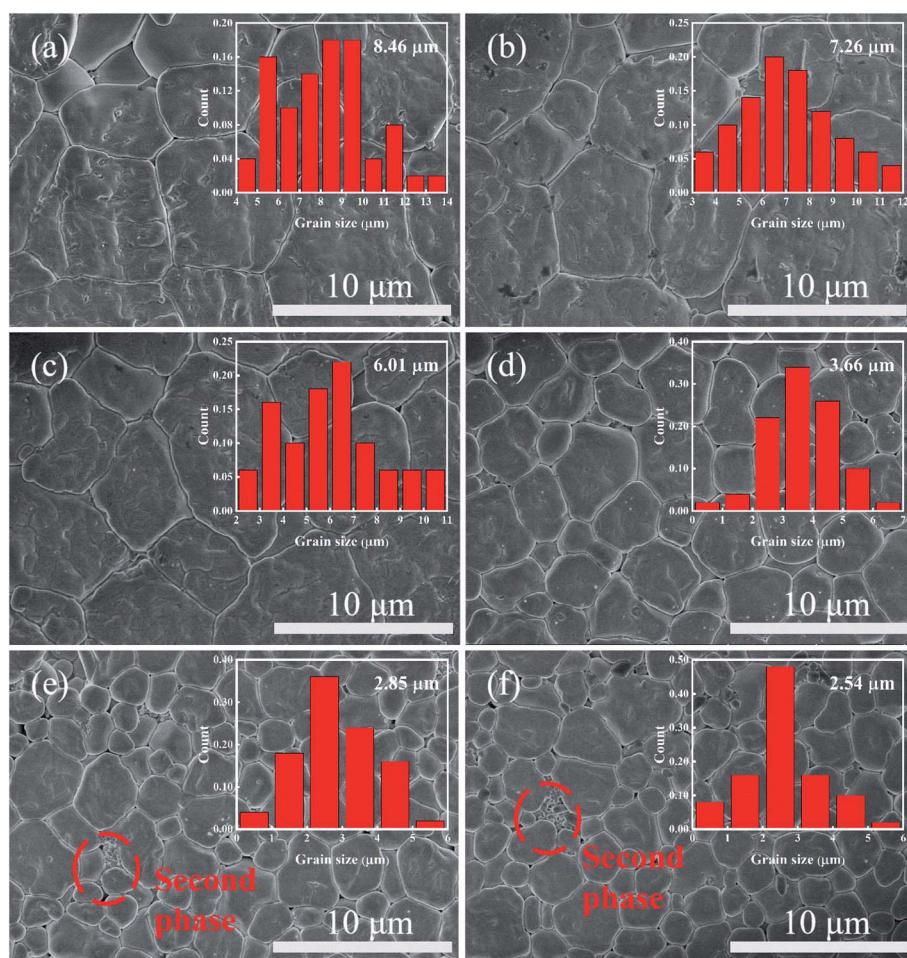


Fig. 3 SEM images of the surface of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics sintered at 1080 °C: (a) 0 mol%, (b) 0.25 mol%, (c) 0.5 mol%, (d) 0.75 mol%, (e) 1.0 mol%, (f) 1.5 mol%. The insets show the statistical data of the grain size of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics.

**Table 1** The variation in the density of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics ( $\rho_1$ : actual density;  $\rho_2$ : theoretical density;  $\rho_r$ : relative density)

$x$ (mol%)	$\rho_1$ (g cm <sup>-3</sup> )	$\rho_2$ (g cm <sup>-3</sup> )	$\rho_r$ (%)
0.00	7.53	7.83	96.17
0.25	7.58	7.82	96.93
0.50	7.64	7.81	97.82
0.75	7.61	7.80	97.56
1.00	7.58	7.79	97.30
1.50	7.52	7.79	96.53

boundary and restrains the grain growth. The results show that Ge<sup>4+</sup> cannot be dissolved into the solid solution.

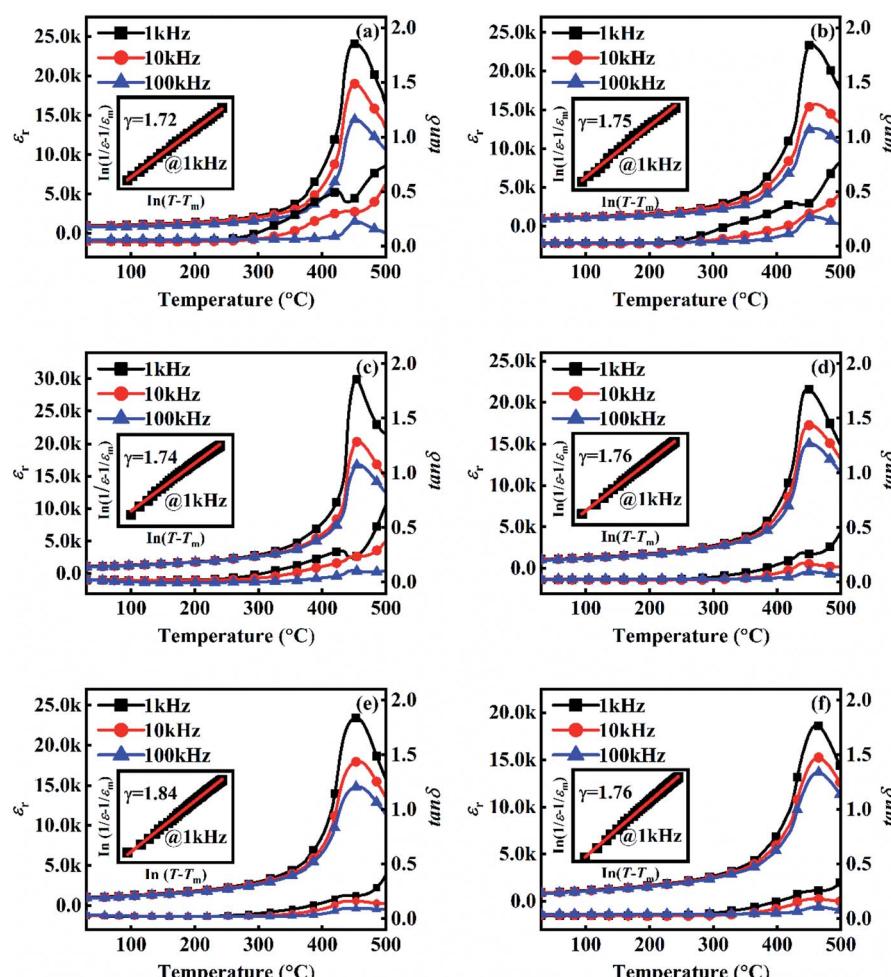
### 3.2 Dielectric properties

Fig. 4 shows the temperature dependence of  $\epsilon_r$  and  $\tan \delta$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics. The temperature of the maximum of dielectric constant ( $T_m$ ) is considered as the Curie temperature ( $T_C$ ).<sup>23</sup> Table 2 summarizes the  $\epsilon_r$ ,  $\tan \delta$  and  $T_C$  of

0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics. The  $T_C$  of 0.36BS–0.64PT ceramics with 0, 0.25, 0.5, 0.75, 1 and 1.5 mol% GeO<sub>2</sub> are 449, 457, 455, 454, 458 and 462 °C, respectively, showing a small change of GeO<sub>2</sub>. The decrease in  $c/a$  indicates that the crystal structure shifts to the rhombohedral phase, and the Curie temperature should be reduced in theory. However, compared with the undoped BS-PT ceramic samples, the introduction of GeO<sub>2</sub> slightly increases the Curie temperature. It is indicated that the addition of GeO<sub>2</sub> is good for stabilizing the Curie temperature.

The values of  $\epsilon_r$  at room temperature increased first and then decreased with the increase in GeO<sub>2</sub>, as shown in Table 2. The maximum  $\epsilon_r$  observed when  $x = 0.5$  mol% was 1280. An increase in the dielectric constant is related to an increase in density. The addition of GeO<sub>2</sub> to 0.36BS–0.64PT ceramics can improve the microstructure of the material. In addition, the GeO<sub>2</sub> doping is beneficial to reducing the dielectric loss.

It can be easily seen that all the samples exhibit a diffuse phase transition. The variation of the diffuse phase transition

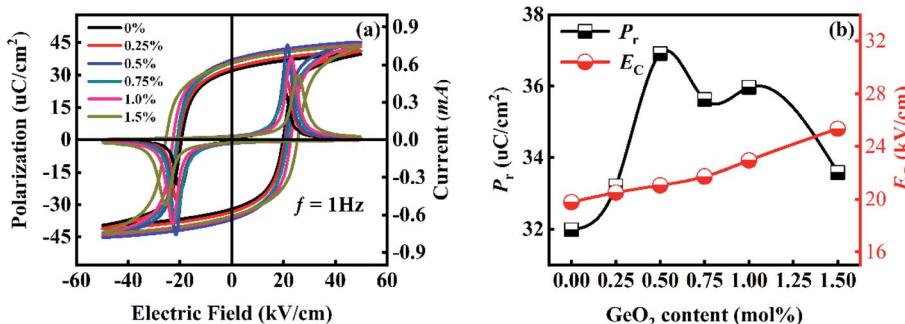


**Fig. 4** Temperature dependence of  $\epsilon_r$  and  $\tan \delta$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics: (a) 0 mol%, (b) 0.25 mol%, (c) 0.5 mol%, (d) 0.75 mol%, (e) 1.0 mol%, (f) 1.5 mol%. The insets show the fitting results using the semi-empirical Curie–Weiss law.



Table 2 Electrical properties of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics at room temperature

$x$ (mol%)	$T_C$ (°C)	$\epsilon_r$ (@1k Hz)	$\tan \delta$ (%)	$E_C$ (kV cm <sup>-1</sup> )	$P_r$ ( $\mu\text{C cm}^{-2}$ )	$d_{33}$ (pC N <sup>-1</sup> )	$k_p$	$Q_m$
0.00	449	1130	3.31	19	32	355	0.49	28
0.25	457	1150	1.47	20	33	370	0.53	75
0.50	455	1280	1.18	21	37	385	0.58	90
0.75	454	1240	1.44	22	36	335	0.55	101
1.00	458	1200	1.71	23	36	305	0.54	115
1.50	462	1070	1.97	25	35	270	0.52	132

Fig. 5 (a)  $P$ - $E$  loops and  $I$ - $E$  loops, (b) the variation of  $P_r$  and  $E_c$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics.

with increase in GeO<sub>2</sub> was explored. The diffuse phase transition can be described by the modified Curie–Weiss law:<sup>32</sup>

$$\ln(1/\epsilon - 1/\epsilon_m) + \ln C = \gamma \ln(T - T_m) \quad (1)$$

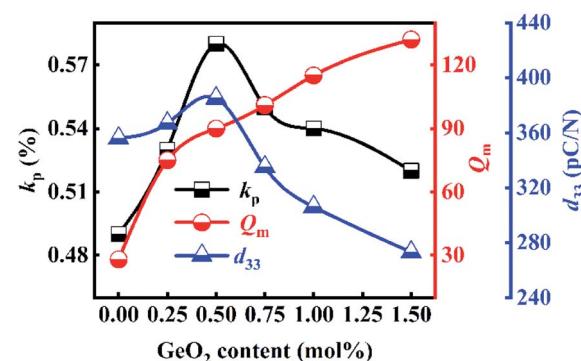
where  $C$  is the Curie Weiss constant,  $\epsilon_m$  is the value of  $\epsilon_r$  at  $T_m$ , and  $\gamma$  is the degree of phase transition diffuses. When  $\gamma$  is equal to 1, it is usually regarded as a normal ferroelectricity. For  $\gamma = 2$ , the relaxor ferroelectric completely diffuses the phase transition. Alternatively, the larger the  $\gamma$  value, the greater the degree of diffuse phase transition. The inset in Fig. 4 shows the fitting curve at 1 kHz of the 0.36BS-0.64PT +  $x$ GeO<sub>2</sub> ceramics, showing that the value of  $\gamma$  for all the ceramic samples is in the range of 1.72–1.84, indicating a strong relaxation behavior. The relaxor behavior is related to the heterogeneity of the solid solution. The complex ions occupy both the A-site (Bi, Pb) and B-site (Ti, Sc, Ga) in the perovskite structure, leading to a disordered distribution of ions.<sup>33</sup> Therefore, the value of  $\gamma$  reflects the degree of heterogeneity. The close values of  $\gamma$  indicate that the GeO<sub>2</sub> addition does not vary the degree of heterogeneity, which is consistent with the stability of the Curie temperature.

### 3.3 Ferroelectric and piezoelectric properties

Fig. 5(a) shows the  $P$ - $E$  loops and  $I$ - $E$  loops of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics measured at room temperature with 1 Hz. According to the  $P$ - $E$  hysteresis loops, the variation of the values of remnant polarization ( $P_r$ ) and coercive field ( $E_c$ ) was obtained, as shown in Fig. 5(b) and Table 2. The  $P_r$  value of the virgin BS-PT ceramics is 32  $\mu\text{C cm}^{-2}$ . The  $P_r$  values increase significantly when a small amount of GeO<sub>2</sub> is added. 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics have the maximum  $P_r$  value (37  $\mu\text{C cm}^{-2}$ ).

cm<sup>-2</sup>). When the GeO<sub>2</sub> added is higher than 0.50 mol%, the value of  $P_r$  decreases slightly because of the glass phase formed by the addition of GeO<sub>2</sub>. The value of  $E_c$  gradually increases as the GeO<sub>2</sub> content increases, indicating the enhanced breakdown strength.

Fig. 6 shows the variation trend of piezoelectric coefficient ( $d_{33}$ ), planar electromechanical coupling factor ( $k_p$ ), and mechanical quality factor ( $Q_m$ ) of the 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics. The detailed data of  $d_{33}$ ,  $k_p$ ,  $Q_m$  are listed in Table 2. As shown in Fig. 6, the  $d_{33}$  and  $k_p$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics increased first and then decreased. The maximum value of  $d_{33}$  and  $k_p$  is 385 pC N<sup>-1</sup>, 58% at  $x = 0.50$  mol%, which is higher than that of the virgin BS-PT ceramics (355 pC N<sup>-1</sup>, 49%). The enhanced piezoelectric performance is attributed to the combined effect of the grain size. The piezoelectric

Fig. 6 The variation of  $k_p$ ,  $Q_m$  and  $d_{33}$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics.

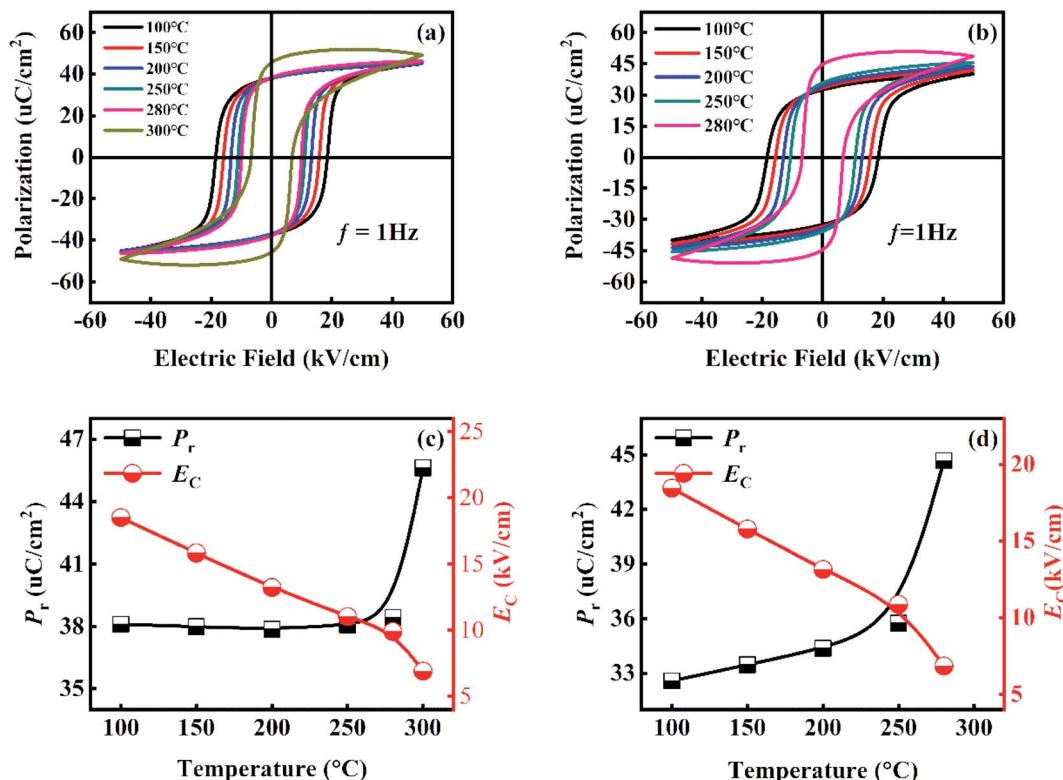


Fig. 7 Temperature dependence of (a and b)  $P$ – $E$  loops, and (c and d)  $P_r$  and  $E_c$  for (a and c) 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> and (b and d) 0.36BS–0.64PT ceramics.

performance decreased as the GeO<sub>2</sub> content increased beyond 0.50 mol%. The addition of GeO<sub>2</sub> can obstruct the movement of the domain and reduce the external contribution to piezoelectricity, resulting in a decrease in  $d_{33}$ .<sup>29</sup> Interestingly, the  $Q_m$  value of ceramic increased monotonically with the increase in the GeO<sub>2</sub> concentration. It is concluded that the addition of GeO<sub>2</sub> can improve the mechanical quality factor, which is consistent with the variation of  $E_c$ .

### 3.4 Thermal stability

Thermal stability is valuable for sensors and actuators in various industrial applications. Fig. 7 shows the temperature dependence of  $P$ – $E$  loops of the 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> and 0.36BS–0.64PT. All curves were measured under an electric field of 50 kV cm<sup>-1</sup> with a frequency of 1 Hz. As the temperature increased, the  $P$ – $E$  loops of 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics still maintained a good rectangular loop at 280 °C without obvious leakage conductance. When the temperature rose to 300 °C, the leakage current was too large to form a saturated  $P$ – $E$  loop confirmed by the abnormal high value of  $P_r$  (see Fig. 7(a)). The variation of the  $P_r$  and  $E_c$  of 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics are shown in Fig. 7(c). The  $E_c$  decreased from 19 kV cm<sup>-1</sup> at 100 °C to 10 kV cm<sup>-1</sup> at 280 °C, while the  $P_r$  remained almost unchanged. The clear leakage conductance appears at 280 °C for 0.36BS–0.64PT ceramics, as shown in Fig. 7(b and d), which is consistent with literature that BS-PT ceramics begin to degrade at 250 °C.<sup>34</sup> The temperature stability

of piezoelectric devices based on BS-PT ceramics is only 250 °C.<sup>35,36</sup> Therefore, it is indicated that the addition of GeO<sub>2</sub> is in favor of the enhancement of the thermal stability of BS-PT ceramics.

To further characterize the temperature stability of the samples, the poling samples were annealed at different temperatures for one hour, and their  $d_{33}$  was measured again at room temperature (Fig. 8). It can be easily seen that the  $d_{33}$  of 0.36BS–0.64PT ceramics starts decreasing above 250 °C.

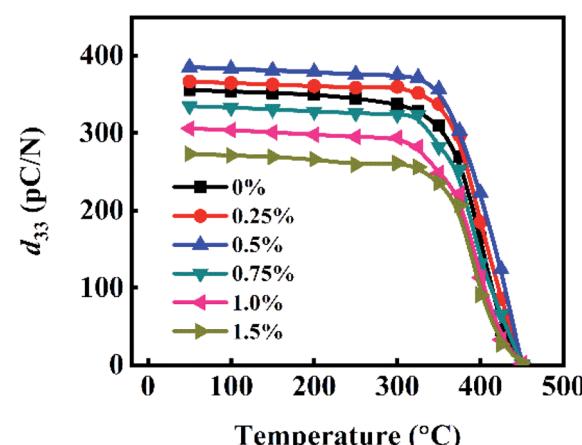


Fig. 8 The thermal stability of  $d_{33}$  of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics.



Compared with 0.36BS–0.64PT ceramics, the 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics exhibit unchanged  $d_{33}$  above 300 °C. This indicates that GeO<sub>2</sub> addition improves the piezoelectric performance of BS-PT ceramics.

## 4. Conclusions

The phase structure, microstructure, dielectric, ferroelectric, and piezoelectric properties of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics were presented. A small amount of GeO<sub>2</sub> can decrease the grain size. With the increase in the GeO<sub>2</sub> content, the values of  $\epsilon_r$ ,  $d_{33}$ ,  $k_p$  increased first and then decreased, while the values of  $E_C$ ,  $Q_m$  increased monotonically. Interestingly, the Curie temperature of 0.36BS–0.64PT +  $x$ GeO<sub>2</sub> ceramics changes only slightly with different contents of GeO<sub>2</sub>. The optimal composition is 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics with  $T_C$  of 455 °C,  $d_{33}$  of 385 pC N<sup>-1</sup>,  $k_p$  of 58%,  $Q_m$  of 90. Compared with 0.36BS–0.64PT ceramics, the enhanced  $Q_m$  and stability of 0.36BS–0.64PT + 0.5 mol%GeO<sub>2</sub> ceramics is due to the addition of GeO<sub>2</sub>. This study provides a paradigm for improving the mechanical quality factor of BS-PT-based ceramics.

## Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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