Review on high temperature piezoelectric ceramics and actuators based on BiScO₃–PbTiO₃ solid solutions

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In recent years, industries in the fields of petrochemical, automotive, and aerospace have expressed their interest in utilizing piezoelectric actuators for high-temperature (HT) operations at the temperature above 150°C. HT piezoelectric ceramics (1 – x) BiScO₃–xPbTiO₃ (BS–PT) are considered as one of the most competitive piezoelectric actuation materials used in the harsh environment because of its high Curie temperature ($T_c = 450^\circ\text{C}$) as well as high piezoelectric coefficient ($d_{33} = 460\text{ pC/N}$) in the MPB composition. This paper summarized the recent progress in HT piezoelectric ceramics and actuators based on BS–PT solid solutions. The properties of BS–PT piezoelectric ceramics and actuators, including driving mechanisms, actuation performances and their applications in HT environment were also discussed.

Keywords: High temperature piezoelectric ceramics; high temperature piezoelectric actuators; BiScO₃–PbTiO₃ solid solutions.

1. Introduction

Piezoelectric actuators have been widely used in adaptive optics, deformable mirrors, camera modules, high resolution positioning stages, and micro-mechanical systems because of their so many merits, such as compact, energy-saving, and capable of generating fine motions ranging from nanometers to millimeters.^{1–4} Recently, piezoelectric actuators have been considered to replace electromagnetic devices to improve the performance of machines. For example, if the piezoelectric actuators were used to replace solenoid valves for more efficient injections in diesel engines, which can further improve fuel efficiency, reduce CO₂ and NO_x emission, and lower engine noise significantly.^{5–9} Such actuators should work effectively under harsh environment with the temperature above 150°C.^{5–9}

Chemical modified lead zirconate titanate (PZT)-based ceramics with the composition near morphotropic phase boundary (MPB) are chosen as piezoelectric active materials, since PZT-based ceramics exhibit excellent piezoelectric performances.^{10–15} However, seriously depoling and ageing problems in PZT-based ceramics at the temperature above 150°C make them unsuitable for high-temperature (HT) actuation operations.^{7,16–18} HT piezoelectric material systems such as GaPO₄ ($T_{\alpha-\beta} = 930^\circ\text{C}$), langasites ($T_c = 1470^\circ\text{C}$) and La₂Ti₂O₇ ($T_c = 1500^\circ\text{C}$) have low piezoelectric coefficients that are not suitable for high performance piezoelectric applications at higher temperature ($> 200^\circ\text{C}$).⁶ Accordingly, the piezoelectric ceramics with both high Curie temperature

($> 400^\circ\text{C}$) and MPB have drawn more attentions because the piezoelectric coefficient d_{ijk} , is enhanced at the MPB region due to the coexistence of multiple phases, such as rhombohedral and tetragonal phases, whose polarization vectors become more readily aligned by an applied electric field. BiScO₃–PbTiO₃ (BS–PT) ceramics exhibit higher Curie temperature ($T_c = 450^\circ\text{C}$) and comparable piezoelectric properties ($d_{33} = 460\text{ pC/N}$), relative to PZT-based ceramics.^{9,19–22} These results indicate BS–PT ceramics were promising candidates for HT piezoelectric devices.

Piezoelectrically generated displacements from a simple single piezoelectric ceramic is quite small ($\sim 3\text{--}6 \times 10^{-4}\text{ }\mu\text{m/V}$).²³ In order to produce big enough displacements, piezoelectric actuators are normally designed into (i) rigid displacement devices, such as multilayered monolith ceramics, ceramic/metal laminated unimorph, bimorph, moonies, cymbal configurations, for which the strain is induced by piezoelectric body under an applied direct current or pulse field,^{24–27} and (ii) resonating displacement devices, such as ultrasonic piezomotors, for which the alternating strain is generated by an alternating current field at the electromechanical resonance frequency.^{28–31} Generally, the conventional piezo-actuators are comprised of piezoelectric ceramics, elastic metal plates as well as polymers; and epoxy resin is usually introduced in these actuators to glue them together. However, at the temperature above 150°C, epoxy resin always becomes “soft”, and is even volatilized. In addition, thermal expansion coefficients for different materials in the same devices are significantly different. Apparently, the

two factors may significantly deteriorate the performance of piezoelectric actuators under HT operation. Therefore, piezoelectric actuators with new working principles should be proposed.

To our best knowledge, it is difficult to find piezoelectric actuators which have good performance at the temperature as high as 200°C in the literatures. Very recently, the piezoelectric parameters of BS-PT ceramics used for the design of actuators have been characterized, and several kinds of HT piezoelectric actuators with new working principle have been proposed in our group.^{32–37} It has been found that these piezoelectric actuators work stably at the temperature as high as 200°C. In this paper, recent progress in HT piezoelectric ceramics and actuators based on BS-PT solid solutions are summarized.

2. Electric Properties of BS-PT Ceramics

Based on the analysis of relationship between perovskite tolerance factor t of Bi(Me)O₃ end member and Curie temperature T_c of MPB compositions for PbTiO₃-Bi(Me)O₃-based solid solutions, Eitel *et al.*^{19–21} predicted that Bi(Me)O₃-PbTiO₃ systems (Me: Sc, In, Y, Yb, etc.) should have higher T_c than that of PZT. Figure 1 shows the phase diagram of BS-PT ceramics with PbTiO₃ content of 50 mol%. A MPB is found at approximately 64 mol% PbTiO₃. BS-PT ceramics with the composition near MPB exhibited higher Curie temperature of 450°C and comparable piezoelectric properties ($d_{33} = 460$ pC/N), relative to PZT-based ceramics.²¹

Recently, it is also found that adding small amount of ABO₃ end members in BS-PT solid solutions, such as Ba(Sr, Ti)O₃, LiNbO₃, LiSbO₃, Pb(Zn_{1/3}Nb_{2/3})O₃, Pb(Mn_{1/3}Nb_{2/3})O₃, and Pb(Sc_{1/2}Nb_{1/2})O₃ are able to increase the piezoelectric constant up to 500 pC/N.^{38–44} However, the ABO₃ end members will decrease the Curie temperature obviously. Wang *et al.* found that the BS-PT ceramics derived from

two-step method has a large d_{33} value of 700 pC/N with slightly decreased Curie temperature of 446°C.^{45,46} The enhanced piezoelectric properties may result from the fine grain, which indicates small domain structure in the BS-PT ceramics. This result implies that “domain engineering” may be an effective way to further improve the piezoelectric properties of BS-PT ceramics.

2.1. Full set of material parameters

To design the piezoelectric devices, full set of elastic, piezoelectric and dielectric parameters of piezoelectric ceramics are needed. The poled piezoelectric ceramics have the symmetry group of ∞m containing 10 independent parameters of five elastic constants, three piezoelectric coefficients and two dielectric permittivities.^{47–49} As it is well known, the rigid piezoelectric actuators are made of “soft” type piezoelectric materials, while the resonating actuators comprise of “hard” type piezoelectric materials. Both “soft” and “hard” piezoelectric materials based on BS-PT ceramics were investigated in the temperature range from room temperature to 500°C.

As shown in Table 1, values of elastic (s_{ij} , c_{ij}), piezoelectric (d_{ij} , e_{ij} , g_{ij} , h_{ij}), dielectric (ϵ_{ij} , β_{ij}), and electromechanical coefficients (k_{ij}) at room temperature of BS-PT ceramics with MPB composition are similar to those of commercial PZT5A ceramics.³² Values of piezoelectric coefficients (d_{ij} , g_{ij}) are higher than those of Mn modified BS-PT ceramics. It is noted that values of d_{33} , d_{15} , k_{15} , d_{33}/d_{31} , g_{33} and Q_m are 441 pC/N, 620 pC/N, 0.71, 3.7, 47.3×10^{-3} Vm/N, and 28, respectively, showing typical characteristics of “soft” piezoelectric ceramics.³² These results indicate that BS-PT ceramics may be suitable for high performance actuators or/sensors.

Table 2 gives the room temperature values of elastic (s_{ij} , c_{ij}), piezoelectric (d_{ij} , e_{ij} , g_{ij} , h_{ij}), dielectric (ϵ_{ij} , β_{ij}), and electromechanical coefficients (k_{ij}) of BS-PT ceramics.⁹ The dielectric, and piezoelectric constants of Mn-modified BS-PT ceramics are lower than those of pure BS-PT ceramics, and comparable to PZT5A ceramics. It is noted that Mn-modified BS-PT ceramics have lower dielectric loss, higher coercive field and higher mechanical quality factor, showing characters of “hard” piezoelectric ceramics. However, the mechanical quality factor of Mn-modified BS-PT ceramics is about one order magnitude lower than those of typical “hard” PZT8 ceramics.

2.2. Temperature dependence of dielectric and piezoelectric properties

Thermal depoling temperature of ferroelectrics determines the usage range of their applications in piezoelectric devices. Figures 2(a)–2(e) exhibit the temperature dependence of electromechanical (k_{ij}), piezoelectric (d_{ij} , g_{ij}), dielectric (ϵ_{ij}), and elastic (s_{ij}) coefficients for BS-PT ceramics.³² It is observed that k_{ij} coefficients increase initially from room

Fig. 1. Phase diagram of $(1-x)$ BiScO₃- x PbTiO₃ ceramics.²⁴

Table 1. Piezoelectric, dielectric, and elastic coefficients of BS-PT ceramics with MPB composition.

Coefficients		Coefficients	
ρ (kg/m ³)	7640	g_{33} (10 ⁻³ Vm/N)	47.3
σ	0.301	g_{15} (10 ⁻³ Vm/N)	39.1
ε_{11}^T	1791	s_{11}^E (10 ⁻¹² m ² /N)	12.59
ε_{33}^T	1051	s_{12}^E (10 ⁻¹² m ² /N)	-3.79
ε_{11}^S	1173	s_{13}^E (10 ⁻¹² m ² /N)	-12.54
ε_{33}^S	732	s_{33}^E (10 ⁻¹² m ² /N)	45.57
$\tan\delta$	0.039	s_{44}^E (10 ⁻¹² m ² /N)	49.30
N_p (Hzm)	1709	s_{66}^E (10 ⁻¹² m ² /N)	32.76
N_T (Hzm)	1591	s_{11}^D (10 ⁻¹² m ² /N)	11.12
N_{31} (Hzm)	1612	s_{12}^D (10 ⁻¹² m ² /N)	-5.24
N_{33} (Hzm)	1195	s_{13}^D (10 ⁻¹² m ² /N)	-6.50
N_{15} (Hzm)	1147	s_{33}^D (10 ⁻¹² m ² /N)	22.92
k_p	0.58	s_{44}^D (10 ⁻¹² m ² /N)	24.45
k_t	0.52	s_{66}^D (10 ⁻¹² m ² /N)	32.76
k_{31}	0.34	c_{11}^E (10 ¹⁰ N/m ²)	29.39
k_{33}	0.70	c_{12}^E (10 ¹⁰ N/m ²)	23.28
k_{15}	0.71	c_{13}^E (10 ¹⁰ N/m ²)	14.50
d_{31} (pC/N)	-117	c_{33}^E (10 ¹⁰ N/m ²)	8.62
d_{33} (pC/N)	441	c_{44}^E (10 ¹⁰ N/m ²)	2.03
d_{15} (pC/N)	620	c_{66}^E (10 ¹⁰ N/m ²)	3.05
d_h (pC/N)	207	c_{11}^D (10 ¹⁰ N/m ²)	30.73
e_{31} (C/m ²)	9.34	c_{12}^D (10 ¹⁰ N/m ²)	24.62
e_{33} (C/m ²)	4.08	c_{13}^D (10 ¹⁰ N/m ²)	13.91
e_{15} (C/m ²)	12.71	c_{33}^D (10 ¹⁰ N/m ²)	11.69
h_{31} (10 ⁸ V/m)	-14.4	c_{44}^D (10 ¹⁰ N/m ²)	4.09
h_{33} (10 ⁸ V/m)	6.28	c_{66}^D (10 ¹⁰ N/m ²)	3.05
h_{15} (10 ⁸ V/m)	12.2	Q_m (disk)	28
g_{31} (10 ⁻³ V*m/N)	-13.7		

Table 2. Piezoelectric, dielectric, and elastic coefficients of Mn-modified BS-PT ceramics.

Coefficients		Coefficients	
ρ (kg/m ³)	7600	g_{33} (10 ⁻³ Vm/N)	23.1
σ	0.301	g_{15} (10 ⁻³ Vm/N)	34.6
ε_{11}^T	1110	s_{11}^E (10 ⁻¹² m ² /N)	12.1
ε_{33}^T	1250	s_{12}^E (10 ⁻¹² m ² /N)	-3.7
ε_{11}^S	695	s_{13}^E (10 ⁻¹² m ² /N)	-5.0
ε_{33}^S	750	s_{33}^E (10 ⁻¹² m ² /N)	14.8
$\tan\delta$	0.01	s_{44}^E (10 ⁻¹² m ² /N)	32.0
N_p (Hzm)	—	s_{66}^E (10 ⁻¹² m ² /N)	31.5
N_T (Hzm)	—	s_{11}^D (10 ⁻¹² m ² /N)	11.2
N_{31} (Hzm)	—	s_{12}^D (10 ⁻¹² m ² /N)	-4.6
N_{33} (Hzm)	—	s_{13}^D (10 ⁻¹² m ² /N)	-2.6
N_{15} (Hzm)	—	s_{33}^D (10 ⁻¹² m ² /N)	8.9
k_p	0.48	s_{44}^D (10 ⁻¹² m ² /N)	20.1
k_t	0.49	s_{66}^D (10 ⁻¹² m ² /N)	31.5
k_{31}	0.28	c_{11}^E (10 ¹⁰ N/m ²)	12.9
k_{33}	0.70	c_{12}^E (10 ¹⁰ N/m ²)	6.5
k_{15}	0.71	c_{13}^E (10 ¹⁰ N/m ²)	6.5
d_{31} (pC/N)	-103	c_{33}^E (10 ¹⁰ N/m ²)	11.0
d_{33} (pC/N)	255	c_{44}^E (10 ¹⁰ N/m ²)	3.1
d_{15} (pC/N)	340	c_{66}^E (10 ¹⁰ N/m ²)	3.2
d_h (pC/N)	49	c_{11}^D (10 ¹⁰ N/m ²)	13.1
e_{31} (C/m ²)	-3.4	c_{12}^D (10 ¹⁰ N/m ²)	6.7
e_{33} (C/m ²)	14.7	c_{13}^D (10 ¹⁰ N/m ²)	5.7
e_{15} (C/m ²)	10.6	c_{33}^D (10 ¹⁰ N/m ²)	14.5
h_{31} (10 ⁸ V/m)	-5.2	c_{44}^D (10 ¹⁰ N/m ²)	5.0
h_{33} (10 ⁸ V/m)	22.1	c_{66}^D (10 ¹⁰ N/m ²)	3.2
h_{15} (10 ⁸ V/m)	17.3	Q_m (disk)	200
g_{31} (10 ⁻³ V*m/N)	-9.3		

temperature to 150–180°C, and then are almost unchanged until the temperature up to 380°C, showing good temperature stability. Figure 2(b) exhibits the temperature dependence of elastic coefficients s_{ij}^E . Generally, these coefficients increase slightly with the rise of the temperature below 350°C, and they go up sharply above this temperature. It is interesting that the value variations of s_{11}^E , $-s_{12}^E$, and $-s_{13}^E$ with temperature are gentler than those of s_{33}^E and s_{44}^E .

As given in Fig. 2(c), it is found that piezoelectric constants d_{ij} of BS-PT ceramics increase with the rise of temperature and reach maximum near the phase transition temperature, and finally drastically fall down. Values of d_{33} , $-d_{31}$, and d_{15} are 1278, 407, and 1775 pC/N, respectively, when the temperature is 380°C. Figure 2(d) shows the temperature dependence of free dielectric constants ε_{ij}^T of BS-PT ceramics. Both ε_{33}^T and ε_{11}^T change slightly with the increasing temperature below 350°C, showing good temperature stability in this region. The temperature-dependent g_{ij} are present in Fig. 2(e). The coefficients of g_{33} , $-g_{31}$, and g_{15} are found to decrease with increasing temperature and become zero near the Curie temperature. These results are similar to those of PZT ceramics, PMN-PT and PIN-PMN-PT crystals. As for ferroelectric materials, the spontaneous

polarization will decrease with increasing temperature.^{50,51} Accordingly, the tendency of coefficients g_{ij} with respect to the temperature is reasonable. However, even at the temperature up to 380°C, values of g_{33} and g_{15} are as high as 21 and 26×10^{-3} Vm/N, respectively. It implies that BS-PT ceramics may be suitable for high performance sensors at high temperature.

As shown in Fig. 3, the temperature-dependent dielectric and piezoelectric constants of Mn modified BS-PT ceramics are compared with those of PZT5A ceramics.⁵² It is observed that the coupling factor stays nearly constant up to 440°C for the modified BS-PT66 material, while the value decreases very quickly when $T > 300^\circ\text{C}$. The temperature coefficient of dielectric constant ($\Delta\varepsilon/\Delta T$) was found to be very flat over the temperature range 25–400°C, which are only 31°C^{-1} and 15°C^{-1} for modified BS-PT ceramics. The piezoelectric coefficients are found to increase with temperature, due to the increase of dielectric constant. Both piezoelectric coefficients and electromechanical coupling factors drop sharply as the samples depolarized, approaching their Curie temperature. It is noted that the temperature stability of dielectric and piezoelectric properties for Mn-modified BS-PT ceramics is better than that of pure BS-PT ceramics.

Fig. 2. The temperature dependence of (a) electromechanical (k_{ij}), (b) elastic (s_{ij}), (c) piezoelectric (d_{ij}), (d) dielectric (ϵ_{ij}), and (e) piezoelectric coefficients (g_{ij}) for BS-PT ceramics.³²

2.3. Important material properties for actuator applications

The regime piezoelectric actuators involve the application of an electric field to a piezoelectric material to produce a strain. For HT piezoelectric actuators, the temperature-dependent strain is the most important concern. Figures 4(a)–4(d) exhibit the temperature-dependent strains of PZT-5H, PZT-4, PZT-8 and BS-PT ceramics under different plus and minus electric fields.³⁴ Although the strain of PZT-5H ceramics is much higher than that of BS-PT ceramics at room temperature, it is clear to see that the strain of PZT-5H ceramics decreases with the rise of the temperature above 100°C due to

its serious depoling problem under the applied electric field. When the temperature rises to 150°C or above, it was difficult to detect a significant strain from PZT-5H ceramics. Furthermore, it is obvious that the HT strain of BS-PT ceramics is larger than those of PZT-4 and PZT-8 ceramics, especially at the temperature above 150°C. At the temperature of 250°C, for example, the strains for BS-PT ceramics under the electric field of ± 7.5 kV/cm are as high as 0.125%, which is 3–4 times higher than those of PZT-4 (0.031%), and PZT-8 ceramics (0.041%). The strains of PZT-4 and PZT-8 ceramics drop more quickly than that of BS-PT ceramics with the rise of temperature above 200°C. Under the electric field of

Fig. 3. The temperature dependence of (a) dielectric (ϵ_{ij}) constant and loss, (b) electromechanical coupling factor and piezoelectric coefficient for Mn-modified BS-PT ceramics.⁵²

± 7.5 kV/cm, it is even difficult to observe detectable strains at the temperature above 250 $^{\circ}\text{C}$ for PZT-4 and PZT-8 ceramics. These measurement results show that BS-PT ceramics possess better HT actuation performances than those of other commercial PZT-based ceramics.

In addition, under each driving electric field, the strains of these piezoelectric ceramics increase with the rise of measuring temperature initially, and then decrease slightly. As it is well known, in a polycrystalline system, the overall piezoelectric responses are from both intrinsic and extrinsic

contributions.^{53,54} The intrinsic piezoelectric response refers to the atomic lattice contributions at the unit cell level, and the extrinsic contributions are from the movement of non-180 $^{\circ}$ domain walls and phase boundaries. The extrinsic responses are usually more important than intrinsic responses. With the increase of temperature, activation energy required for the domains changing from one stable state to another becomes lower. Accordingly, the piezoelectric performance is improved at the lower temperature. The decrease in strain at higher temperature and electric field may be attributed to the

Fig. 4. (a)–(d) Temperature-dependent strains of PZT5H, PZT4, PZT8 and BS-PT ceramics under different electric field.³⁴

Fig. 5. (a) Vibration velocity of Mn-modified BS-PT ceramics as a function of applied electric field. (b) Temperature rise of Mn-modified BS-PT ceramics as a function of vibration velocity.³³

fact that both electric and mechanical loss of piezoelectric ceramics become worse with the temperature and electric field further increase, which then restricts their piezoelectric effect.

Resonating actuators work under the electromechanical resonance frequency. Therefore, the vibration velocity and generated heat under the alternative current electric field with resonate frequencies are paid much attentions. The combination of high T_c with a piezoelectric coefficient d_{33} of ~ 450 pC/N, which is comparable to those of commercial PZT ceramics, make them promising candidates for HT piezoelectric applications. However, the mechanical quality factor Q_m and dielectric dissipation factor $\tan\delta$ of BS-PT ceramics are 28% and 4% (10^3 Hz), respectively, indicating that they may generate too much heat while they work under resonance frequency as power electronic devices.^{32,33} It was reported that Mn modification was an effective way to increase the mechanical quality factor Q_m and decrease the dielectric dissipation factor $\tan\delta$ of BS-PT ceramics simultaneously. The mechanical quality factor Q_m of Mn-modified BS-PT is about seven times higher than that of pure BS-PT ceramics.³²

Figures 5(a) and 5(b) display the vibration velocity v_o (rms value) as a function of the electric field (rms value) and temperature rise as a function of vibration velocity of Mn-modified BS-PT ceramics, respectively.³⁷ It is observed that the vibration velocity increases with both the electric field and Mn content. For the compositions with 3 and 4 mol% Mn content, the vibration velocity of Mn-modified BS-PT ceramics increases rapidly with the electric field initially, and then become nearly saturated with further increasing electric field. This saturation may be due to a decrease in mechanical quality factor Q_m with increasing electric field above a critical vibration level, as well as heat generation. Under the electric field of 7 V/mm (rms value), the vibration velocity of Mn-modified BS-PT ceramics with Mn content of 3 and 4 mol% reaches up to about 0.9 m/s, which is around four times that of unmodified BS-PT ceramics and 2.2 times that of the

conventional hard PZT ceramics with a value of about 0.40 m/s (rms value) at higher electric drive level.^{55,56}

As shown in Fig. 5(b), the temperature rise of Mn-modified BS-PT ceramics decrease dramatically with Mn content, reflecting that Mn modification is an effective way to decrease the generated heat from BS-PT ceramics driven under resonance frequency. From the practical perspective, the maximum vibration velocity was defined as the v_o which generates $\Delta T = 20$ K. It is observed that the maximum vibration velocity of Mn-modified BS-PT ceramics reaches the maximum value at Mn content of 3 mol%, which is as high as 1.05 m/s. This is much superior to those of PZT-PZM (0.72 m/s) and PZT (0.4 m/s) ceramics. Considering the Curie temperature ($T_c = 431^\circ\text{C}$), piezoelectric properties ($d_{33} = 317$ pC/N) and maximum vibration velocity ($v_o = 1.05$ m/s), 0.36BS-0.64PTM ceramics has potential for HT and high power piezoelectric device applications.

3. HT Piezoelectric Actuators Based on BS-PT Ceramics

3.1. Multilayer piezoelectric actuators

Multilayer piezoelectric actuators (MPAs) have attracted considerable attentions because of their large generative force, high efficiency, quick response, and no electromagnetic noise, relative to the electromagnetic actuators.⁵⁷⁻⁵⁹ MPAs are usually made of PZT ceramics due to their excellent piezoelectric properties. Recently, MPAs have been used to replace electromagnetic solenoid valves for more efficient injections in diesel engines, which can further improve fuel efficiency, reduce CO_2 and NO_x emission, and lower engine noise significantly. The device is required to work stably as high as 150°C . However, the conventional PZT ceramic materials have serious depoling and aging problems near the temperature of 150°C . The combination of high T_c and comparable piezoelectric coefficient d_{33} relative to PZT-based ceramics make BS-PT ceramics promising candidates for HT actuation applications.

Fig. 6. (a) Photograph and (b) electrode configuration of the MPA-BS-PT (the arrow denotes the direction of polarization).³⁴

The photograph and electrode configuration of the actuator fabricated using BS-PT ceramics are given in Figs. 6(a) and 6(b), respectively.³⁴ As can be seen, the multilayer actuator is composed of 10 layers, and each one is $12.5 \times 12.5 \times 1 \text{ mm}^3$ in size. The internal electrode is only $12.5 \times 10 \text{ mm}^2$ in sizes for electric insulating of adjacent layers. The thickness of adhesive used in the actuator is about 0.15 mm. The multilayer actuator was poled in an oil bath under an electric field of 40 kV/cm at 120°C for 30 min. The multilayer actuators formed from BS-PT ceramics were called MPA-BS-PT.

Figures 7(a)–7(d) show the measured displacements of MPA-BS-PT actuators and a single layer BS-PT ceramic at different typical temperatures under $\pm 7.5 \text{ kV/cm}$ electric field

with triangular voltage waveform. It is found that the displacement shape of the MPA-BS-PT at different temperatures is also typical triangular, indicating that the displacement of the actuator is coincident with the driving electric field.

Taking data from Figs. 7(a)–7(d), the strains of the MPA-BS-PT-G and single layer BS-PT ceramic are presented in Fig. 8. The strains and displacements of the MPA-BS-PT increase significantly with the temperature from room temperature up to 150°C, and then decrease slightly. The trend of strain with temperature for the MPA-BS-PT prepared by simple glue method is similar to that of BS-PT single layer ceramics. Under $\pm 7.5 \text{ kV/cm}$ electric field, the strain and displacement of the MPA-BS-PT at 200°C are as high as 0.115% and $11.5 \mu\text{m}$, respectively. As can be seen, the strain value of MPA-BS-PT is about 80% of that of a single layer ceramic in the temperature range from 25°C up to 200°C. The strain loss may be attributed to the silicate cement between ceramic layers, which restricts the strain of BS-PT piezoelectric ceramics. However, the loss due to the cement is acceptable. For example, it is only about 18% under 200°C. This result is similar to that of a co-fired PZN-PZT/Ag multilayer actuator prepared by tape-casting method.^{48,49}

Fig. 7. Dynamic displacement responses of the MPA-BS-PT actuators and single layer BS-PT ceramics at different temperatures.³⁴

Fig. 8. Strains of the MPA-BS-PT and single layer BS-PT ceramics under $\pm 7.5 \text{ kV/cm}$ at different temperatures.³⁴

Fig. 9. Strain versus electric field for MPA-BS-PT under ± 7.5 kV/cm at different temperatures.³⁴

Strains versus an applied electric field of the MPA-BS-PT under ± 7.5 kV/cm at different temperatures are shown in Fig. 9. The MPA-BS-PT shows good linear behavior in the temperature range. The hysteresis loops are observed for MPA-BS-PT actuator at low frequency, which has been observed in other piezoelectric multilayer actuators. This may result from the irreversible domains. Moreover, it is interesting that the shape of loops under different temperatures is symmetric, which is useful for device applications.

3.2. Shear-bending piezoelectric actuators

Generally, conventional piezo-actuators are comprised of piezoelectric ceramics, elastic metal plates, and polymers, and epoxy resin is usually used to glue these components together. However, the epoxy resin may become soft while cracks may generate due to the different thermal expansion coefficients of different materials at high temperature. Therefore, a simple single ceramic actuator without the use of epoxy resin may be more suitable for HT actuators.

The photograph of the ring-shaped BS-PT ceramic actuator is shown in Fig. 10³⁵. The ceramics were poled along

Fig. 10. Photograph of ring-shaped HT BS-PT ceramic actuator.³⁵

Fig. 11. Piezoelectrically excited shear-bending deformation of the ring-shape actuator calculated by ANSYS FEM software.³⁵

the radial direction at a temperature 15°C higher than the Curie point under an electric field of 1 kV/cm for 30 min, and were then cooled to room temperature in air inside the furnace while the poling electric field was increased to 3 kV/cm. Next, the silver electrodes were removed from the samples, and gold electrodes were sputtered onto both top and bottom surfaces of the ceramic rings.

The generated deformations and displacements of the ring-shaped actuator under the applied electric fields were analyzed and simulated. ANSYS Finite Element Method (FEM) software was used to calculate the simulated deformation, shown in Fig. 11. It was observed that when a positive (negative) direct current voltage is applied to the piezoelectric ring-plate along the thickness direction, an axial-symmetric shear strain is generated according to the piezoelectric relationship: $S_5 = d_{15}E_1$, where d_{15} is the piezoelectric shear strain constant and E_1 is the applied electric field. This shear strain results in radial expansion (or contraction) of the top surface of the ring-plate while its bottom surface contracts (or expands) in the radial direction, leading to an upward (or downward) bending deformation of the entire actuator. This displacement mechanism has not been previously reported. The simulated displacement at the center of the ring-shaped actuator without a load under an applied electric field of 7.5 kV/cm is about $6\text{ }\mu\text{m}$.

On the assumption of a shear-bending deformation, the displacement D at the center of the ring-shaped actuator along the thickness direction is also estimated by the following formula:

$$D = (R_1 - R_2)d_{15}E_1, \quad (1)$$

where R_1 and R_2 are the radii of the outer and inner rings, respectively. However, the calculated result is about 33% higher than that of the FEM simulation. This is because the restriction of shear-strain in the circumference direction of the ring-shaped actuator is neglected in Eq. (1).

Finally, the displacement responses of the BS-PT ceramic ring-shaped actuator to the applied electric field were measured at different temperatures. Figure 12 shows dynamic displacement responses of the ring-shaped actuator at different temperatures of 25°C , 50°C , 150°C , and 200°C under a load of 2.5 N and an applied triangular waveform electric voltage (corresponding to an electric field of ± 7.5 kV/cm). It can be seen that the generated displacement from the ring-shaped actuator was about $8\text{ }\mu\text{m}$ at room temperature, reaching about $20\text{ }\mu\text{m}$ at 200°C . Note that the displacement corresponding to 7.5 kV/cm is only $4\text{ }\mu\text{m}$, which is less than the FEM simulated displacement value

Fig. 12. Dynamic displacement response of the ring-shaped actuator at different temperatures.³⁵

(about 6 μm). This is attributed to measurement being carried out under a load of 2.5 N.

As a comparison, Fig. 13 shows the displacements of the ring-shaped actuator and a single layer BS-PT ceramic plate with the same thickness. It is noted that the displacement of the ring-shaped actuator at ± 7.5 kV/cm is about 11–14 times that of the single layer BS-PT ceramic pellet in the temperature range 25°C to 200°C. It can also be seen from Fig. 13 that the displacements of both actuators initially increase with temperature, reach a maximum near 150°C, and then decrease slightly at 200°C.

Displacement shapes of the ring-shaped actuator versus electric field under ± 7.5 kV/cm at different temperatures are given in Fig. 14. It is clear that the BS-PT actuator shows good linear behavior with low hysteresis in the temperature range measured. Further observation confirmed that the positive displacement of the ring-shaped actuator is equal to the negative displacement, which indicates that the shear-bending deformation generated from the ring-shaped actuator has reversible displacement. This feature is important for device applications.

Fig. 13. Displacement of ring-shaped actuator and single layer BS-PT ceramic at different temperatures.³⁵

Fig. 14. Strain versus electric field for ring-shaped actuator under ± 7.5 kV/cm at different temperatures.³⁵

3.3. L_1 - B_2 piezoelectric motor

One of the typical designs for a miniature piezoelectric motor is to use a double-mode piezoelectric rectangular plate vibrator in which the first longitudinal (L_1) and the second bending (B_2) vibration modes are excited synchronously.^{60–63} The resultant motion of the two vibration modes is a desired elliptic motion, which is then used to drive a contacted slider into linear motion via friction force. Figure 15(a) shows the schematic drawing of the piezoelectric motor structure, and Fig. 15(b) shows a photo of the piezoelectric motor.⁶³ The vibrator in the piezoelectric motor is a simple piezoelectric Mn-modified BS-PT ceramic plate (16.35 mm(L) $\times$ 4.7 mm (W) $\times$ 2.0 mm(T)), and its top electrode is divided into four parts, acting as two channel terminals for two-phase driving of the vibrator; the bottom electrode of the vibrator is a whole area, acting as ground electrode. The piezoelectric Mn-modified BS-PT ceramic plate is polarized along its thickness direction. Thus, elliptical motions at frictional driving tip can be generated due to resultant motion of L_1 and B_2 double modes. Because the driving tip of the piezoelectric vibrator is pressed against the slider, these microscopic elliptic motions finally cause a macroscopic linear motion of the slider through frictional forces.

Figures 16(a)–16(d) show the load-speed and load-efficiency relationships at different temperatures, while the driving voltage is fixed to 100 V_{p-p}.⁶³ It can be seen that the velocity decreases almost linearly with load. At each temperature, there is an optimal load corresponded to a maximum efficiency. At 90°C, the linear motor shows a maximum driving force of 1.18 N. At the temperature of 200°C, although both the maximum driving force and motion speed of the linear motor decreases to only 0.35 N and 41.7 mm/s, respectively, it still work steady. The decrease in driving force or velocity is mainly attributed to the vibration mode splitting, large dielectric or mechanical loss of the BS-PT-Mn ceramic at 200°C. In addition, the plastic mount softening at 200°C temperature may have some effects on motion stability of the linear motor.

Fig. 15. Structure of the L_1 - B_2 linear motor. (a) Schematic of the linear motor. (b) Photo of the linear motor.⁶³

Fig. 16. Load characteristics of the linear motor at different temperatures.⁶³

3.4. B_1 - B_1 piezoelectric motor

As mentioned above, the load and speed performances of the L_1 - B_2 piezoelectric actuator dropped dramatically at the temperature above 150°C. Investigations reflected that the resonance frequency difference between L_1 and B_2 modes of

the piezoelectric actuator enlarged with the rise of the temperature. Therefore, new type of linear piezoelectric motor should be proposed. Figure 17(a) shows the schematic drawing of the piezoelectric actuator, and Fig. 17(b) shows its photo.³⁶ The ultrasonic actuator consists of a single Mn-BS-PT square-plate with sizes of 10 mm × 10 mm × 1 mm. Silver

(a)

(b)

Fig. 17. (a) Schematic of the linear actuator. (b) Photo of the linear actuator.³⁶

electrodes were patterned on both sides of the Mn-BS-PT ceramic plate. The top electrode was divided into four parts (a, b, c, and d region) for voltage drive and the bottom electrode was a uniform area for ground. The polarization direction of regions a and c were opposite to that of regions b and d. Regions a and b were connected as CH1 while regions

c and d were connected as CH2 for two phase voltages input. A frictional tip was glued at the center of one side of the square plate for frictional driving. By applying one pair of orthogonal ac voltage with the B₁-mode resonance frequency to CH1 and CH2 of the actuator, an elliptical motion was excited at the frictional tip of the actuator, leading to the slider moving linearly through friction force between the tip and slider. By controlling the phase shift ($\pm 90^\circ$) of the two voltages, the moving direction can be reversed.

Figures 18(a) and 18(b) show the temperature-dependent spectrums of the Mn-BS-PT actuator measured by an impedance analyzer (HP 4294A, Agilent Technologies Inc., USA). It was found that only one resonance frequency was found for both CH1 and CH2 in this frequency range (110–140 kHz), indicating that two pure B₁ vibration modes had been excited. The two shapes of impedance and phase spectra with frequency in the whole measured temperature range for CH1 and CH2 are almost coincided. This result implies that Mn-BS-PT actuator operating in B₁-B₁ mode exhibits better temperature stability than actuators operating in L₁-B₂ mode.

Figures 19(a)–19(f) compare the relationship between loads and speed as well as efficiency at different temperatures under the electric field of 100 V_{p-p}. The speed of Mn-BS-PT actuator decrease almost linearly with load, and there is an optimal load corresponding to a maximum efficiency. The maximum speed, load and efficiency increase with the temperature up to about 100°C initially which are 220 mm/s, 0.45 N and 4%, respectively, and then decrease slightly. It is noted that the maximum speed, load and efficiency under 200°C is comparable to those under 150°C.

In order to evaluate the temperature stability of B₁-B₁ mode piezoelectric actuator, the normalized load, speed and efficiency relative to those at 100°C under different temperatures are compared in Fig. 20. For comparison, the normalized load, speed and efficiency for a L₁-B₂ piezoelectric actuator under different temperatures are also given. The variations of normalized load and speed with temperature

(a)

(b)

Fig. 18. (a) Impedance and (b) phase spectra of the actuator at different temperatures.³⁶

Fig. 19. Load characteristics of the Mn-BS-PT actuator at different temperatures.³⁶

from 50 °C to 200 °C for the B_1 - B_1 mode actuator is only about 12% and 14%, respectively, which are about 25% of those for an actuator operating in L_1 - B_2 mode. Note that at 200 °C, the efficiency of the B_1 - B_1 mode actuator has a large drop due to HT loss of Mn-BS-PT ceramic material. However, it is still much higher than that of a L_1 - B_2 mode actuator. Clearly, compared with L_1 - B_2 mode, Mn-BS-PT actuator

operating in B_1 - B_1 mode shows much better HT driving performance.

4. Conclusion

In summary, HT piezoelectric ceramics and actuators based on BS-PT solid are briefly reviewed. The full set of temperature-dependent material parameters for both “soft” and “hard” type BS-PT ceramics have been characterized in the temperature range from 25 °C to 500 °C. The dielectric, piezoelectric and elastic properties of BS-PT ceramics stable up to 350 °C. Furthermore, the HT strain of BS-PT ceramics is larger than those of PZT-5A, PZT-4 and PZT-8 ceramics, especially at the temperature above 150 °C. At the temperature of 250 °C, for example, the strains for BS-PT ceramics under the electric field of ± 7.5 kV/cm are as high as 0.125%, which is 3–4 times higher than those of PZT-4 (0.031%), and PZT-8 ceramics (0.041%). The vibration velocity of Mn-modified BS-PT ceramics with Mn content of 3 and 4 mol% reaches up to about 0.9 m/s at the electric field of only 7 V/mm (rms value), which is around four times greater than that of unmodified BS-PT ceramics and 2.2 times greater than that of the conventional hard PZT ceramics with a value of about 0.40 m/s (rms value) at higher electric drive level. These results indicate that BS-PT ceramics are suitable materials used for HT piezoelectric applications. However, the high cost of raw material Sc_2O_3 may hinder their commercial applications. Further work may focus on exploring cheap HT piezoelectric materials.

Fig. 20. The variation of maximum load, speed and efficiency with the increasing temperature for Mn-BS-PT piezoelectric actuator operating in B_1 - B_1 and L_1 - B_2 modes.³⁶

The multilayer piezoelectric and shear-bending actuators based on BS-PT ceramics have been studied. It is found that the actuators work stably in the temperature range from 25°C to 200°C. The displacement of the shear-bending actuator at ± 7.5 kV/cm is about 11–14 times higher than that of the single layer BS-PT ceramic pellet in the temperature range 25°C to 200°C. However, the displacement of multilayer piezoelectric and shear-bending actuators under various frequencies need to be further investigated. The B_1 - B_1 mode piezoelectric actuator work stabler than that of L_1 - B_2 mode piezoelectric actuator in the temperature range from 25°C to 200°C. However, the HT efficiency and speed should be further improved.

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