Influence of temperature on the electrical characteristic parameters and dynamic electro-mechanical behaviour of PZT-5H

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Abstract In the field of ammunition, piezoelectric ceramics are increasingly used as part of impact fuze systems. It is an urgent problem to improve the low-temperature storage and low-temperature environment adaptability of piezoelectric ceramic power supply. In order to study the dynamic mechanical and electrical response characteristics of PZT-5H at $-40$ to $25$ °C, the static electrical parameters of PZT-5H at different temperatures were measured, and the Hopkinson bar experiments at different temperatures and impact velocities were carried out by using the self-established low-temperature mechanical and electrical test system. The variation of resistivity and relative permittivity of PZT-5H from $-40$ to $25$ °C is obtained by experiment, and the relaxation time of PZT-5H is obtained according to Debye’s theory. Combined with the observation of scanning electron microscope (SEM), the influence mechanism of porosity and crack on the electrical output characteristics of piezoelectric ceramics was obtained. The mechanism of the freezing effect of piezoelectric ceramics at low temperatures is obtained through space charge theory analysis. The variation of the dynamic piezoelectric voltage constant with temperature and time in the elastic region measured by the experiment is clarified.

1 Introduction

Piezoelectric ceramics are widely used in high dielectric constant capacitors, piezoelectric transducers, high displacement piezoelectric actuators, nanopiezoelectric sensors, optical limiters and other device materials and have become the core of aerospace, electronic information, ammunition engineering and other fields.

Jaffe et al. [1] had made PZT ferroelectric ceramics for the first time. With the maturity of preparation technology, many scholars had carried out a lot of experimental researches on the properties of PZT series ceramics. Mckinney et al. [2] studied the piezoelectric properties of mixed titanate ceramics at different pressures (0–1000 kg/cm$^2$) in the range of $-25$ °C to 37.5 °C. The results show that the piezoelectric constant increases with the increase in

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temperature, but there is an obvious phase transition near 5 °C. Zhang et al. [3] designed an experimental method to directly and quantitatively measure the domain wall and intrinsic contribution of piezoelectric and dielectric response of ferroelectric materials based on the fact that the motion of domain wall does not produce volume change. By using this method, the contribution of domain wall and intrinsic part of PZT series ceramics and its relationship with temperature were evaluated. The motion of the domain wall induces the piezoelectric and dielectric properties of ceramics. Noheda et al. [4] compared the phase transition temperature of PZT ferroelectric ceramics with 91–97% zirconium content under two conditions of heating and cooling and studied the hysteresis phenomenon of phase transition of ferroelectric materials with different loading ways and different zirconium titanium ratio. It is found that there is a discontinuous phase transition near the Curie temperature in the two doped ceramics by means of the dielectric constant and loss data. Hooker [5] measured the dielectric, ferroelectric and piezoelectric properties of PZT-4, PZT-5A, PZT-5H and PLZT-9/65/35 in the temperature range of −150 to 250 °C. The results showed that PZT-4 was relatively stable in the temperature range of −150 °C to 25 °C, and PZT-5H had better piezoelectric properties at room temperature. The physical mechanism of the influence of low temperature on the dielectric, ferroelectric and piezoelectric properties of PZT-5H was not clarified. Panda [6] carried out thermal shock fatigue test on PZT-5 piezoelectric ceramics. Each side of the sample keeps a constant temperature, but there is a large temperature difference between the two sides, resulting in a temperature gradient in the material, so as to simulate the effect of thermal shock on ferroelectric ceramics. It was observed that the thermal stress is very sensitive to the temperature distribution on the top surface and maximum near the periphery of the top surface. Andryushina et al. [7] revealed the relationship between the relaxation process of PbTi0.495Zr0.505O3 solid solution and the change of structural characteristics in the temperature range of 100–300 K. This study only explained the change rule of temperature on the structural characteristics of PZT-based ceramics and did not discuss its electrical characteristics and electro-mechanical coupling characteristics.

With the study of temperature dependence of piezoelectric materials, the piezoelectric properties of piezoelectric ceramics at different temperatures under quasi-static conditions have been widely concerned. Xue et al. [8] studied the basic mechanical properties and fracture mechanism of zirconia-based structural ceramics, alumina ceramics and nonoxide ceramics such as silicon nitride and silicon carbide at low temperature. However, the mechanical properties and fracture mechanism of structural ceramics were only analysed under quasi-static condition, and the dynamic properties at low temperature were not studied. Lan et al. [9] revealed the change rule of phase structure and electrical properties of dense PZT95/5 ferroelectric ceramics at low temperature, but did not further explain the microchange of phase structure and electrical properties of dense PZT95/5 under low temperature. Anand et al. [10] proposed a temperature-dependent micromechanical model based on internal energy to evaluate the ferroelectric and ferroelastic behaviour of PZT wafers, extracted macrostate variables (residual polarization, residual strain, maximum polarization and maximum strain, etc.) and discussed them as a function of temperature. Cao et al. [11] applied the phenomenological theory of ferroelectrics and the response of electric domain to alternating electric field and put forward a model of the hysteresis freezing effect of the domain in the measurement of hysteresis loop and explained reasonably the phenomenon of polarization degradation at low temperature. Belovickis et al. [12] studied the dielectric spectrum, ferroelectric and piezoelectric properties of P(VDF-TrFE)/BPZT in the temperature range of 150–420 K and analysed the relationship between the effective dielectric constant of the composite and the filler concentration by Lichtenecker’s method.
The service environment of piezoelectric materials is often very complex, and temperature, strain rate and other factors are the key to study their mechanical and electrical properties. Therefore, the research on the dynamic mechanical and electrical properties of piezoelectric materials at different temperatures has gradually become the focus of scholars all over the world. Tang et al. [13–17] have done a lot of research on the dynamic mechanical and electrical properties of PZT-5H and obtained the mechanical response and piezoelectric properties of PZT-5H at different strain rates. They also evaluated the electrical output characteristics of laminated piezoelectric ceramics under overload conditions and studied the influence mechanism of internal cracks on the discharge characteristics. Flannigan et al. [18] found that the energy index of PZT was strongly dependent on temperature and evolved into a superposition value of about 2.0 with the change of temperature. Wang et al. [19] studied the temperature dependence of PZT-5H piezoelectric properties. The experimental results showed that the piezoelectric strain coefficients of PZT-5H change linearly with temperature, and the decrease in temperature leads to the decrease in the measured piezoelectric coefficients. The internal mechanical stresses are the main mechanism of PZT-5H ferroelectric fatigue, and a set of enhanced constitutive relation, it can be used to extrapolate some nonlinear characteristics to other temperature regions are established.

In conclusion, there are many researchers on the electrostatic properties of piezoelectric ceramics at room temperature, but the dynamic mechanical–electrical response characteristics of piezoelectric ceramics at low temperature are yet not clear. In this paper, the resistivity and relative permittivity of PZT-5H in the temperature range of $-40$ to $25\, ^\circ\text{C}$ are measured, and Hopkinson pressure bar experiments are performed for PZT-5H at the temperature of $-40\, ^\circ\text{C}$, $-25\, ^\circ\text{C}$, $-10\, ^\circ\text{C}$, $0\, ^\circ\text{C}$ and $25\, ^\circ\text{C}$ and different impact velocities. Besides, the fracture and the electro-mechanical response characteristics of PZT-5H at different temperatures, and impact velocities are analysed based on the theory of one-dimensional elastic stress wave propagation and microscopic observation.

2 Materials and methods

2.1 Materials

The PZT-5H (Pb$_{1.0}$[Zr$_{0.49}$Ti$_{0.46}$ (La$_{0.25}$Sb$_{0.5}$Fe$_{0.25}$)$_{0.05}$]O$_3$) cylindrical piezoelectric ceramic with the diameter and height of $\Phi 14 \times 3\, \text{mm}^2$ produced by Yuhai Electronic Ceramics Co., Ltd, China, is used in the experiment. Figure 1 shows a photograph of the PZT-5H ceramic. Wherein the mark ‘+’ on the ceramic surface is the positive electrode, and the polarization direction is along the thickness direction. The basic performance parameters of PZT-5H at $25\, ^\circ\text{C}$ provided by Yuhai Electronic Ceramics Co., Ltd, are listed in Table 1.

2.2 Experimental method

PZT-5H static tests have been conducted in TH2832 LCR digital tester of Changzhou Tonghui Electronic Co., Ltd, China. The low-temperature PZT-5H experiments at different impact velocities were carried out on the separated Hopkinson pressure bar experimental system in the Key Laboratory of Transient Physical Mechanics and Energy Conversion Materials of Liaoning Province, China. Figure 2 is the schematic diagram of the experimental system.

The conductive copper foil (the thickness is 20 $\mu\text{m}$) is used to connect the positive and negative electrodes of the ceramic. Polyimide film with a thickness of 20 $\mu\text{m}$ was used as the insulating material between the specimen and the bar. Studies have shown that the
influence of the foils with the thickness of 50 µm on one-dimensional wave propagation in rods is negligible [20]. The output voltage at both ends of the electrode is collected by the voltage probe acquisition system, and the direction of the collected voltage is consistent with the polarization direction. The time history curve of the voltage pulse is recorded by the oscilloscope.

The diameter of the bars used in the SHPB system is 16 mm, and the material is aluminium alloy. The length of the transmitting bar is 200 mm, and the length of the incident and the transmitting bars is 1200 mm, respectively. A low-temperature system is installed at the specimen position of SHPB device. The system consists of cavity, liquid nitrogen tank, pipe and control valve. After flowing through the chamber, the liquid nitrogen flows out to the outside to reduce the internal temperature and form a low-temperature environment. The specimen and insulating layer are placed inside the cavity. The temperature in the tank is controlled by controlling the flow rate of liquid nitrogen, and the temperature sensor is used to monitor the temperature of the specimen. The temperature change rate of the specimen should not exceed 3 °C/min to ensure the constant internal temperature of the specimen. In
order to reduce the influence of temperature on the bar, the incident bar and the transmission bar are placed outside the cavity during the cooling process. When the temperature of the specimen reaches the predetermined temperature, keep the constant temperature for 15 min. Finally, the incident rod and the transmission rod are embedded in the cavity and clamped to the specimen, and the experiment is completed in the shortest possible time. In Fig. 2, the enlarged part of the red circle is the schematic diagram of the cooling process of the sample, while the original part is the schematic diagram of the experimental state.

According to one-dimensional elastic stress wave theory [21], the strain rate $\dot{\varepsilon}$, strain $\varepsilon$ and stress $\sigma$ of the specimen can be calculated directly under the assumption of one-dimensional stress loading and uniform distribution of stress and strain along its length

$$\dot{\varepsilon} = \frac{c_0}{l_0} (\varepsilon_1 - \varepsilon_2 - \varepsilon_3)$$  \hspace{1cm} (1)

$$\varepsilon = \frac{c_0}{l_0} \int_0^t (\varepsilon_1 - \varepsilon_2 - \varepsilon_3) \, d\tau$$  \hspace{1cm} (2)

$$\sigma = \frac{AE}{2A_0} (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)$$  \hspace{1cm} (3)

In formulas (1)–(3), $c_0$ is the elastic wave velocity in the bar, $\varepsilon_1$, $\varepsilon_2$ and $\varepsilon_3$ are the strains in the bar corresponding to the incident wave, the reflected wave and the transmitted wave when they propagate independently. $l_0$ and $A_0$ are the original length and cross-sectional area of the specimen, respectively. $A$ and $E$ are the cross-sectional area and elastic modulus of the bar, respectively. The duration of the stress pulse on the specimen is equal to the time of the wave going back and forth once in the impact bar [21], which can be expressed as

$$t = \frac{2L}{c_0}$$  \hspace{1cm} (4)

In formula (4), $L$ is the length of impact bar. It can be known that the loading frequency of the specimen is $\omega_0 = 1/l_0 = 17.75$ kHZ.

3 The influence of temperature on electrostatic characteristic parameters of PZT-5H

3.1 Resistivity

The resistivities of PZT-5H at different temperatures are measured by TH2832 LCR digital tester. Figure 3 shows the spectrum of PZT-5H resistivity at different temperatures. Figure 4 is the relationship between resistivity and temperature of PZT-5H at loading frequency $\omega_0$.

The piezoelectric crystal is an impedance component with resonant frequency $\omega_r$ and anti-resonant frequency $\omega_a$, when the loading frequency on the piezoelectric crystal is equal to its resonance frequency $\omega_r$, the piezoelectric crystal will resonate. At this time, the impedance angle is the maximum value in the negative direction and the resistance is the minimum value of the impedance angle. When the loading frequency increases to the frequency with the impedance angle of zero, the resistance has a peak value. When the loading frequency of the piezoelectric crystal increases to the anti-resonance frequency $\omega_a$, the impedance angle increases to the maximum value of the forward direction, and the resistance is the maximum value of the impedance angle.

As can be seen from Fig. 3, at the temperature range of $-40$ °C to $25$ °C, the resonant frequency of PZT-5H is $147.359$ kHz, and the anti-resonant frequency is $163.931$ kHz. The influence of the frequency between $\omega_r$ and $\omega_a$ on the resistivity of PZT-5H is obvious and the peak appears. Figure 4 shows that the resistivity of PZT-5H decreases with the increase
in temperature at this frequency, but the decrease in resistivity above 0 °C is steeper than that below 0 °C.

In theory, the intrinsic conductance of electrons as carriers and the nonintrinsic conductance of ions in dielectric materials, ion clusters and space charges as carriers determine the resistivity of materials [22]. Conductivity $\sigma_e$, the number $n$ and mobility of conduction electrons $u$ per unit volume are satisfied with

$$\sigma_e = \frac{1}{\rho} = neu$$  \hspace{1cm} (5)
Fig. 5 The relative permittivity spectrum of PZT-5H at different temperatures

At room temperature (25 °C), the carrier is mainly excited by electrons from full band to conduction band, that is, the intrinsic excitation of piezoelectric ceramics. At low temperatures, the carrier is mainly excited by electrons from the impurity donor to the conduction band, that is, the impurity excitation of the piezoelectric ceramics. The intrinsic excitation and impurity excitation of PZT-5H can increase the conduction electron with the increase in temperature. However, the energy difference produced by intrinsic excitation is much larger than that produced by impurity excitation [22]. Therefore, the change trend of intrinsic excitation with temperature is more steep, which is consistent with the conclusion in the literature [5].

3.2 Dielectric properties

Dielectric property is the most basic physical property of piezoelectric ceramics. Because of the loss in piezoelectric ceramic measurement, the relative permittivity $\varepsilon_r$ of piezoelectric ceramics can be expressed in the form of complex number as [23]

$$\varepsilon_r^*(\omega) = \varepsilon'_r(\omega) - j\varepsilon''_r(\omega)$$  \hspace{1cm} (6)

In formula (6), $\varepsilon'_r(\omega)$ is the real part of relative complex permittivity under alternating electric field, and $\varepsilon''_r(\omega)$ is the imaginary part of the relative complex permittivity, representing the dielectric loss. Figure 5 shows the relative permittivity spectrum of PZT-5H at different temperatures. Figure 6 shows the dielectric loss spectrum of PZT-5H at different temperatures.

As can be seen from Fig. 5, in the temperature range of 40–25 °C, the dielectric constant of PZT-5H has obvious differences near the vibration frequency $\omega_1$ [$\varepsilon'_r(\omega_1) = 0$ and $\omega_1$ is the maximum point], and resonance absorption occurs [24], which indicates that the energy dissipation of elastic domains in the electric field is mainly near $\omega_1$. 
Dielectric loss refers to the phenomenon that the dielectric is heated by the external electric field due to the part of consumption of the electric energy. The tangent value of dielectric loss angle can usually be expressed as

$$\tan \delta(\omega) = \frac{\varepsilon''_r}{\varepsilon'_r}$$

(7)

Dielectric loss is generally in the form of polarization loss, leakage loss, ionization loss and structure loss. Polarization loss is closely related to frequency, and structure loss is closely related to the internal tightness of the medium at low temperature. Under the action of alternating electric field, the relation between dielectric behaviour and frequency of ceramics is revealed according to Debye’s Eqs. (8)–(10)

$$\varepsilon'_r(\omega) = \varepsilon_{r\infty} + \frac{\varepsilon_{rs} - \varepsilon_{r\infty}}{1 + \omega^2 \tau^2}$$

(8)

$$\varepsilon''_r(\omega) = \frac{(\varepsilon_{rs} - \varepsilon_{r\infty}) \omega \tau}{1 + \omega^2 \tau^2}$$

(9)

$$\tan \delta = \frac{(\varepsilon_{rs} - \varepsilon_{r\infty}) \omega \tau}{\varepsilon_{rs} + \varepsilon_{r\infty} \omega^2 \tau^2}$$

(10)

where $\varepsilon_{rs}$ is the relative dielectric constant under static state, $\varepsilon_{r\infty}$ is the relative dielectric constant under optical frequency, $\tau$ is the relaxation time. It can be seen from Figs. 5 and 6 combined with formula (8)–(10), when the applied frequency is very low ($\omega \to 0$), the polarization energy can keep up with the change of the electric field, that is to say, all forms of polarization can be completely established and $\varepsilon'_r$ reaches the maximum values without loss. With the increase in frequency, the relaxation polarization cannot keep up with the change of electric field frequency $\omega_1$, and resulting in the dielectric constant decreases and $\varepsilon''_r$ and $\tan \delta$ increase, which results in dielectric loss.

When the condition is $0.01 < \omega \tau < 100$, the dielectric loss of piezoelectric ceramics is generated along with the energy loss, where $\tan \delta$ reaches peak value when $\omega$ is satisfied with $\omega=1/\tau$ or $\omega=\sqrt{\varepsilon_{rs}/\varepsilon_{r\infty}/\tau}$. When the frequency is very high, the relaxation polarization is
Table 2 The relaxation time $\tau$ of PZT-5H at different temperatures

| $T$ (°C) | −40 | −30 | −20 | −10 | 0  | 25 |
|----------|-----|-----|-----|-----|----|----|
| $\tau$ (µs) | 8.154 | 8.128 | 8.102 | 8.076 | 8.050 | 8.024 |

Fig. 7 The curves of relative permittivity $\varepsilon'_r$, dielectric loss and temperature at loading frequency $\omega_0$. 

not enough to establish and has no contribution to the dielectric constant. At this time, the dielectric constant is determined only by the displacement polarization. When the condition is $\varepsilon'_r \rightarrow \varepsilon_{\infty}$, it tends to the minimum value, and the $\varepsilon''_r$ and tan $\delta$ decrease. When the conditions are $\omega \rightarrow \infty$, $\varepsilon''_r \rightarrow 0$ and tan $\delta \rightarrow 0$, there is no dielectric loss.

Among them, relaxation time $\tau$ is a constant independent of time but related to temperature, which means that $\varepsilon'_r$, $\varepsilon''_r$ and tan $\delta$ are also closely related to temperature. According to the peak point of tan $\delta$ in Fig. 6, the relaxation time $\tau$ of PZT-5H at different temperatures can be deduced. Table 2 shows the relaxation time $\tau$ of PZT-5H at different temperatures.

Based on Eqs. (8)–(10), the relaxation time can be expressed as

$$\ln \tau = c + \omega / KT$$

(11)

In Eq. (11), $K$ and $C$ are diffusion coefficient and constant. With the increase in temperature, the relaxation time decreases exponentially. The Debye’s theory hypothesis shows that molecular thermal motion is the main cause of dipole inversion. The action of electric field only changes the direction of dipole turning to electric field. Figure 7 shows the change curve of the relative dielectric constant $\varepsilon'_r$ and dielectric loss of PZT-5H with temperature under the loading frequency $\omega_0$. 

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At room temperature, the relative permittivity of PZT-5H is 3199.3. With the decrease in temperature, the relative permittivity curve decreases to 2306.1 at the temperature of −40 °C. Compared with the temperature of 25 °C, the relative permittivity of PZT-5H decreases by 27.92% at the temperature of −40 °C, but the corresponding loss increased from 0.0156 at the temperature of 25 °C to 0.03 at the temperature of −40 °C. In Fig. 7, the relative permittivity of PZT-5H decreases continuously without mutation, i.e. sudden change, in the process of temperature decreasing and the loss has no peak value, indicating that PZT-5H has no phase transition in this temperature range [25]. The results are consistent with that of PZT95/5 ferroelectric ceramics in reference [9].

In the normal coordinate system, the relative permittivity of $N$-square lattice ceramics is related to the lattice vibration frequency. Anderson [26] pointed out that the effective Hamiltonian can be used to qualitatively explain the dielectric anomaly. When the propagation vector is $\beta = 0$, the effective Hamiltonian of the potential energy of the lattice optical oscillator is related to the coefficient of thermal expansion. According to the Slater-Devonshire theory [26], using the relationship between relative permittivity and lattice vibration frequency and the equilibrium condition of potential energy effective Hamiltonian of lattice oscillator, combining with the relative permittivity in the permittivity equation $\alpha = (\varepsilon_0 \varepsilon_r - 1)/4\pi (\varepsilon_0 = 8.85 \times 10^{-12} \text{ F/m})$ is the vacuum permittivity), it can be obtained that the change of relative permittivity and temperature is related to the coefficient of thermal expansion, i.e. to the effect of thermal expansion [27], the low temperature leads to no more intense thermal motion. The smaller the expansion effect of the lattice decreases with the temperature, the closer the lattice is, the more difficult the domain rotation is, resulting in the decrease in dielectric constant with decreasing temperature.

4 Influence of temperature on the dynamic response characteristics of PZT-5H

At different temperatures (−40 °C, −25 °C, −10 °C, 0 °C and 25 °C), PZT-5H impact tests are carried out at three different impact speeds (2.4 m/s, 3.4 m/s and 4.5 m/s (the specimen breaks at this time)).

4.1 Influence of temperature and impact load on deformation process of PZT-5H

Figure 8 shows the stress and strain curve of PZT-5H at different impact speeds at the temperature of 0 °C.

By observing the recovered post impact specimen, it is known that when the impact velocity is 2.4 m/s, PZT-5H does not occur macrofracture and has no obvious change. When the impact velocity is 4.5 m/s, the linear elastic deformation starts from point “a” to point “b” and then enters the nonelastic stage. The yield stage is in the c–d segment, and the yield point “d” is in the segment, and then entering the plastic hardening stage of point “e” (the fracture strength is 36.19 MPa). At this time, the deformation of the piezoelectric ceramics is straight to the complete fracture. Figure 9 shows the fracture patterns on the surface of piezoelectric ceramics after impact.

It can be seen from Fig. 9 that the piezoelectric ceramics fracture along the main cracks I and II; then, the cracks bifurcate and form cracks III and IV, forming penetrating cracks along the polarization direction. Figure 10 is a partial SEM micrographs of the section along the thickness direction of the fracture at the red arrow in Fig. 9 piezoelectric ceramics.
Fig. 8 The stress and strain curve of PZT-5H at different impact speeds at the temperature of 0 °C

Fig. 9 The fracture patterns on the surface of piezoelectric ceramics after impact

Fig. 10 SEM micrographs piezoelectric ceramics a 500×, b 5000×, c 5000×

Figure 10 shows that PZT-5H is a cubic crystal structure. Figure 11 shows the yield strength at impact speeds of 2.4 m/s and 3.4 m/s, and the relationship between the fracture strength and temperature at impact speeds of 4.5 m/s.

It can be seen from Fig. 11 that the temperature of piezoelectric ceramics decreases from the temperature of 25–0 °C, and the yield and fracture strengths decrease with the decrease...
The yield strength at impact speeds of 2.4 m/s and 3.4 m/s, and the relationship between the fracture strength and temperature at impact speeds of 4.5 m/s at temperature. With the continuous decrease in temperature, the yield and fracture strengths of piezoelectric ceramics increase gradually and reach the stable at the temperature of $-30^\circ$C, and the temperature of $-25^\circ$C is 26.94% higher than that at the temperature of 25 $^\circ$C. According to the fracture morphology of ceramics in Figs. 9 and 10, the increase in steady-state fracture toughness at low temperature may be due to the contribution of phase transformation toughening component [8].

4.2 Influence of temperature on open-circuit discharge characteristics of PZT-5H

Considering the electric effect of piezoelectric ceramics, the electric performance of low-temperature piezoelectric ceramics is tested. The output voltage induced by the temperature change of piezoelectric ceramics is small, and the magnitude difference between the output voltage caused by temperature change and the output voltage of piezoelectric effect is 3–4 orders, so the electric effect can be ignored.

4.2.1 Electrical characteristics

When the piezoelectric ceramics are polarized along the polarization direction by the external force, the charge adsorbed on the two electrodes of the piezoelectric ceramics is released. When the external force is unloaded, the piezoelectric ceramics return to the uncharged state. The charge density is directly proportional to the applied force, which is called the positive piezoelectric effect. The piezoelectric equation can be expressed as

$$Q = d_{33}F$$

where $Q$ is the amount of released charge, $d_{33}$ is the piezoelectric strain constant in the polarization direction, and $F$ is the external force along the polarization direction. Piezoelectric
Fig. 12 The time history curves of stress and output voltage at the temperature of 25 °C. a The impact velocity 2.4 m/s. b The impact velocity 4.5 m/s

Fig. 13 The time history curves of stress and output voltage at the temperature of −40 °C. a The impact velocity 2.4 m/s. b The impact velocity 4.5 m/s

ceramics can be equivalent to capacitance; then, there is \( Q = CU \), where \( U \) is the output voltage, and \( C \) is the capacitance of piezoelectric ceramics. According to the expression of capacitance \( C = \varepsilon_0 \varepsilon_r A_0 / l_0 \), the relationship can be expressed as

\[
U = \frac{Q l_0}{\varepsilon_0 \varepsilon_r A_0} \tag{13}
\]

where \( A_0 \) is the cross-sectional area of the specimen, and \( l_0 \) is the thickness of the specimen.

Substituting Eq. (12) into Eq. (13), the relationship can be expressed as

\[
U = g_{33} l_0 \sigma \tag{14}
\]

In formula (14), \( g_{33} \) is the voltage piezoelectric constant in the polarization direction.

Figures 12 and 13 show the time history curves of stress and output voltage at the temperatures of 25 °C and −40 °C with impact velocities of 2.4 m/s and 4.5 m/s, respectively.

From Figs. 12 and 13, it can be seen that the output voltage of both ends of the piezoelectric ceramic is changed with the applied stress in the linear region at the temperatures of −40 °C and 25 °C. When the piezoelectric ceramic breaks, the voltage drops suddenly, and there are two different peaks in the voltage time history curve, and the second voltage peak is smaller than the first voltage peak, while the difference of the voltage peak before and after the temperature of −40 °C, fracture is larger than that at the temperature of 25 °C. Piezoelectric ceramic, as a porous material, impurity containing heterogeneous material,
Fig. 14  a  The SEM micrograph of ceramics (500×).  b  The SEM micrograph in fracture section of ceramics (5×).  c  Energy spectrum of EDS spot 7 of the SEM micrograph in fracture section of ceramics (5×).  d  Energy spectrum of EDS spot 8 the SEM micrograph in fractures section of ceramics (5×)
its internal microhole defects and impurity defects will affect the macroscopic discharge, and the temperature will change the internal structure of piezoelectric ceramics, thereby increasing the peak voltage difference. Figure 14 shows the SEM micrographs and the energy spectrum of the piezoelectric ceramics, which Fig. 14a shows the defects on the surface of the piezoelectric ceramics (without electrode), Fig. 14b shows the SEM micrograph in fracture section of ceramics with EDS spots 1–8 is the energy spectrum analysis spots, and Fig. 14c and d is the energy spectrum of EDS spot 7 and EDS spot 8, respectively.
In fact, the dielectric constant of the air or ionized gas inside the piezoelectric ceramics is limited, but piezoelectric ceramics can withstand higher electric field strength, so it has a certain conductivity. Mcmeeking [28] and Dunn [29] used the dielectric ellipse model to describe the defects. The long axis and short axis of the ellipse are 2a and 2b, respectively. The dielectric constants of the ellipse and the matrix were $\varepsilon_f$ and $\varepsilon_m$, respectively. The conductivity of the defects is described by the electric field permeability parameter $\kappa$

$$\kappa = \frac{a\varepsilon_f}{b\varepsilon_m}$$  \hspace{1cm} (15)

The porosity defects shown in Fig. 14a can be regarded as ellipses along the polarization direction. The dielectric constants of the interior and the substrate of the ellipse are the dielectric constants of the air at the temperature of 25 °C, respectively, and the long axis and short axis of the ellipse are measured according to the scale to obtain $\kappa = 3.54 \times 10^{-4}$. In this paper, the conductivity of piezoelectric ceramics at the temperature of 25 °C is measured to be $\sigma_e = 9.64 \times 10^{-6} \text{ } \Omega^{-1} \text{ } \text{m}^{-1}$. Comparing the values of $\kappa$ and $\sigma_e$, it can be found that pore defects and crack gaps will affect the voltage output of piezoelectric ceramics, and the
influence degree is related to pore size, pore number and crack gap size. The reduction of temperature makes the grains inside the piezoelectric ceramics shrink, and the size of the pores increases, which increases the output voltage difference.

Figure 14b shows the SEM micrograph in fracture section of ceramics with EDS spots 1–8 is the energy spectrum analysis spots, and Fig. 14c and d is the energy spectrum of EDS spot 7 and EDS spot 8, respectively. In the practical operation of energy spectrum analysis, because the ceramic is a poor conductor, an additional conductive thin-layer material needs to be sputtered on the inner surface of the measured ceramic (the material used in this paper is gold), so the proportion of Au element in Fig. 14c and d is the most. The energy spectrum analysis shows that there are C, K, Na, Ne, Kr, Si and other elements in the piezoelectric ceramics, which indicates that there are impurities in the piezoelectric ceramics.

There is also a discharge phenomenon of piezoelectric ceramics excited by impurities. The electric field formed by the impact makes the charging temperature of the ceramic impurities reach the critical value and discharge, and the further decrease in the temperature affects the voltage output of the piezoelectric ceramics.

4.2.2 The freezing effect

At room temperature, when the piezoelectric ceramics are polarized by an external force along the polarization direction, the charge absorbed by both ends of the electrode is released, and the voltage returns to zero potential as the external force unloading. Due to the ferroelectric of piezoelectric ceramics, when the piezoelectric ceramics break, the voltage will increase reversely to the opposite number of voltage peak value caused by external force. Figures 15 and 16 show that time history curves of output voltage with impact velocity of 3.4 m/s and 4.5 m/s at the temperatures of −25 °C and 25 °C, respectively, only the piezoelectric ceramic is damaged at the temperature of 25 °C, and the voltage at both ends of piezoelectric ceramic will not return to zero potential after the piezoelectric ceramic is discharged. However, in
the case of $-25^\circ C$, there is a phenomenon that the voltage at both ends of the piezoelectric ceramic does not return to zero potential and increases reversely after discharge, including the impact at the temperature of $0^\circ C$ and the impact velocities of 3.4 m/s and 4.5 m/s (that is, the piezoelectric ceramic does not break after impact), respectively. This is related to the rotation of domain in piezoelectric ceramics. In this paper, after unloading at low temperature, the output voltage of unbroken piezoelectric ceramics does not return to zero potential, but reversely increases to the opposite number of voltage peaks, which is called freezing effect.

According to the space charge theory of Okazaki [30], the space charge in the lattice is mostly vacancy and atom, and the space charge generated by polarization generally exists near the grain boundary or domain wall, as shown in Fig. 17.

The discharge of piezoelectric ceramics is composed of the movement of charge in the lattice, and the movement of charge is realized through the movement of vacancies and interstitial atoms in the lattice. The movement of atoms jumps from one lattice position to another, while the jumping of vacancy and interstitial atoms depends on thermal fluctuation [23], so it is closely related to temperature. Taking the gap atom as an example, the gap atom is in a lattice position with a minimum relative potential energy, and there is a maximum potential energy, i.e. potential barrier, between the gap atom and the adjacent gap. The gap atom generally vibrates in the vicinity of the minimum potential energy. The gap atom must cross the barrier to jump the adjacent gap. According to the statistics of the thermal fluctuation probability, the jump rate of the gap atom shows that the movement of the gap atom increases exponentially with the increase in temperature [23]. According to the similar analysis, the vacancy motion can also get similar results.

It can be seen from Fig. 17 that the electric field generated by the impact pressure of piezoelectric ceramics has the opposite effect on the space charge layer in the grain and the domain, thus hindering the domain rotation, the grains of piezoelectric ceramics shrink at low temperature, the space charge layer increases, and the domain wall cannot keep up with the electric field change caused by the impact pressure, which makes it more difficult to rotate. Therefore, at low temperature, the impact pressure of piezoelectric ceramics produces freezing effect without fracture.

The effect of grain size on piezoelectric properties is greater than that on dielectric properties. PZT-5H piezoelectric properties are better at room temperature than at low temperature, which is consistent with the experimental results in the literature [5].

### 4.2.3 Electrical output characteristics

When piezoelectric ceramics work in linear region, due to electric field strength $E = U/l_0$, the relationship can be expressed as

$$E = g_{33}\sigma$$

(16)

Figure 18 shows the stress-time curve at the impact velocity of 3.4 m/s and the temperature of $0^\circ C$, and the electric field strength time history curve at the impact velocity of 3.4 m/s and different temperatures. In the linear region of the figure, the relationship between the piezoelectric voltage coefficient $g_{33}$ and the temperature at different impact speeds is derived from Eq. (16), as shown in Fig. 19. At the same impact velocity, the piezoelectric voltage coefficient increases with the decrease in temperature. At the temperature of $25^\circ C$, there is no significant difference in the piezoelectric voltage coefficient at different impact velocities. At low temperature, the impact velocity has significant influence on the piezoelectric voltage coefficient.
The linear region of the electric field intensity time curve of PZT-5H under stress and different temperatures in Fig. 18 is derived from Eq. (14). The relationship between the dynamic piezoelectric voltage coefficient and temperature of PZT-5H under different impact speeds is shown in Fig. 19. When the ceramic is not broken (impact velocity 2.4 m/s and 3.4 m/s), the piezoelectric voltage coefficient increases with the decrease in temperature under the condition of constant impact velocity. When the ceramic fracture occurs (impact velocity is 4.5 m/s), the electric field strength suddenly decreases at −25 °C. This is due to the decrease in the change rate of electric field intensity in the linear region. When the temperature is constant, the dynamic piezoelectric voltage coefficient changes with the impact velocity. The influence of impact velocity on piezoelectric voltage coefficient increases with the decrease in temperature. However, the impact stress increases with the increase in impact velocity.

According to the above experimental results, the main contributions to the change of dynamic piezoelectric voltage coefficient are temperature and dynamic factors [17] (impact speed, strain rate, whether ceramic fracture occurs, etc.). Due to the coupling effect of the mechanical and temperature environments, there is no good method to decouple and analyse their individual influence on the dynamic piezoelectric voltage constant. This will be the focus of our future research.

5 Conclusion

In this paper, the static electrical parameters of PZT-5H were measured at the temperature of −40 to 25 °C, and the mechanical test system of liquid nitrogen cooling/Hopkinson piezoelectric bar and the voltage output test system of piezoelectric ceramics were established. The stress load and electrical output characteristics of piezoelectric ceramics were measured at different temperatures and impact speeds. The results show that the electrical properties of piezoelectric ceramics at low temperature are lower than that at room temperature, but the mechanical properties of PZT-5H can still maintain a certain level or even have a significant improvement trend at low temperature. Compared with the significant effect of impact on the discharge performance of piezoelectric ceramics, the effect of temperature on the discharge performance is irregular, but it cannot be ignored. In practical engineering application, the influence of the two factors on the discharge performance of piezoelectric ceramics should be comprehensively evaluated. At the same time, it is urgent to establish the mathematical models of stress, strain rate, temperature and piezoelectric constant.

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Declarations

Conflict of interest The authors declare that we have no conflict of interest. The data that support the findings of this study are available from the corresponding author upon reasonable request.
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