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Electron emission from lead lanthanum zirconate titanate ferroelectric cathodes

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Abstract

This work concentrated on studying the relationship between electron emission and materials properties. Ferroelectric (PLZT 7/65/35) and paraelectric (PLZT 12/65/35) compositions of lead lanthanum zirconate titanate (PLZT) system were chosen to study for their widely different ferroelectric and dielectric properties. The experimental results proved the wrong of the spontaneous polarization switching model which based on the ferroelectric properties of the cathode. The mechanism of electron emission from PLZT ceramics was ascertained to field emission and surface plasma emission. The emission current depends slightly on the ferroelectric properties and depends significantly on the dielectric constant.

Keywords: C. Dielectric properties; C. Ferroelectric properties; D. PLZT; E. Nuclear applications

1. Introduction

Electron emission (J_c as high as 100 A/cm^2) from ferroelectric ceramics subjected to a high voltage pulse has received considerable attention since 1988 [1]. There are a number of potential applications of this novel cathode ranging from electron guns [2] to triggering gas switches [3]. Ferroelectrics cathodes have many advantages, such as room temperature and low vacuum operations, high repetition rates, control of the emission and most important, high emission current densities to be useful for high-power microwave applications [2–4].

There is still, however, controversy surrounding the nature of the electron emission process. A commonly accepted ferroelectric emission model is polarization switching. A ferroelectric material that has been polarized will have the positive charges of the dipoles oriented toward the ferroelectric surface between the grid electrodes. The resulting net positive charge on the surface is compensated by electrons to preserve charge neutrality. When fast reversal of the polarization occurs, the negative charges of the ferroelectric dipoles orient toward the surface, which leads to a rapid buildup of a repulsive electrostatic force. This coulombic repulsion is thought to be responsible for the ejection of electrons from the ferroelectric surface [5–9]. However, recent experimental investigations have shown that high electron emission current was observed in both the paraelectric phase ferroelectrics and ferroelectrics above Curie temperature. It was pointed out that the electron emission from ferroelectrics is a plasma-assisted emission [10–13]. Controversies surrounding the nature of the electron emission hindered the progress of the novel ferroelectric cathode.

Although, electron emission from ferroelectrics has been studied for almost 20 years, not much data about materials properties, especially polarization switching and piezoelectric constant together with electron emission has been published, thus far. In this study, we focused on studying the relationship between electron emission and materials properties. Parallel study of the emission characteristics of ferroelectric (PLZT 7/65/35) and paraelectric (PLZT 12/65/35) compositions of PLZT system would give a clear path to see if ferroelectric properties play an important role in the electron emission. A better understanding of the phenomena may open an avenue for successful application of this electron emission.

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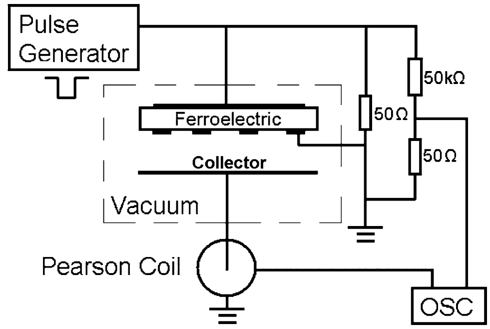

Fig. 1. Experimental setup.

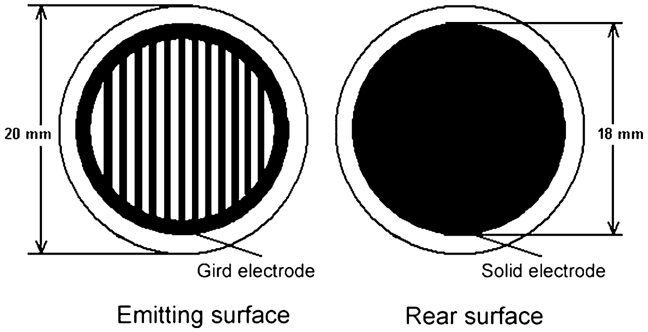

Fig. 3. Sketch of the ferroelectric cathode.

2. Experimental procedure

2.1. Ferroelectric emission measurement system

The experimental setup is illustrated in Fig. 1. The pulse generator was comprised of a fast high voltage switch and a DC power supplier (ZGF-30/5, Peking Fengyuan Tec.). This combination can generate high voltage (<10 kV) pulses of 250 ns with a fast rise time (<20 ns) under a repetitive rate of ~1 Hz. Ferroelectric cathodes were set in the vacuum chamber with the grid electrode facing the electron collector (a flat graphite disk). The collector was kept at the ground potential. The distance between the sample and the collector is 5 mm. The triggering pulse was applied to the rear electrode of samples, while the grid electrode was kept at the ground potential. A Pearson current monitor (Model 6595, Pearson Electronics Inc.) and a digital oscilloscope (TDS 2012, Tektronix Inc.) were used to detect the emission current. In order to minimize electrical noise, all connector cables were shielded and made as short as possible. All studies were performed in a vacuum of 1×10^{-5} Torr and at room temperature of 25 °C.

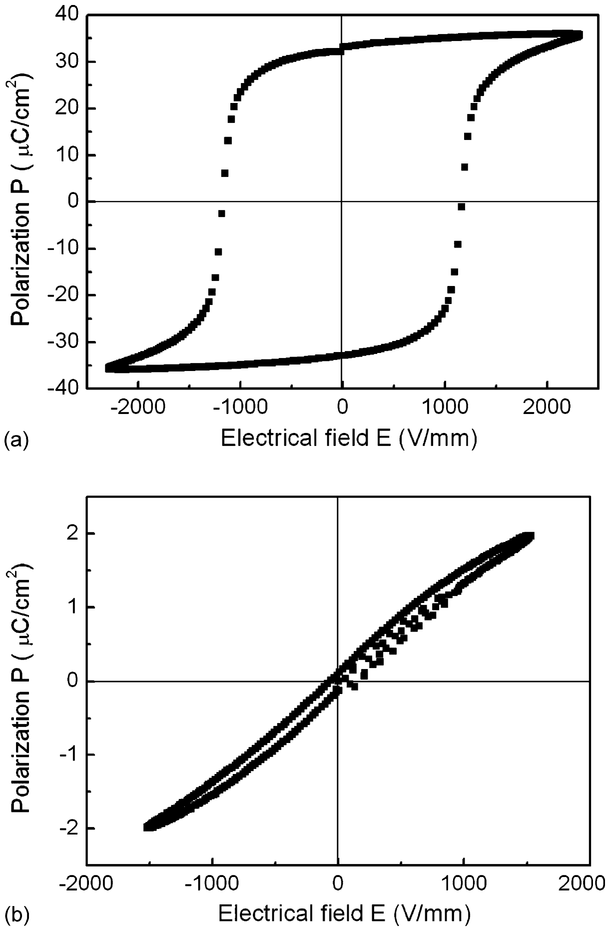

Fig. 2. Hysteresis loops of (a) PLZT 7/65/35 and (b) 12/65/35 ceramics.

2.2. Ferroelectric sample preparation

Ferroelectric (PLZT 7/65/35) and paraelectric (PLZT 12/65/35) compositions of lead lanthanum zirconate titanate (PLZT) system were used for comparison. The dielectric constant of the two compositions is about 1600 and 3100, respectively. Fig. 2 shows the hysteresis loops of the PLZT 7/65/35 and PLZT 12/65/35 ceramics at room temperature (25 °C). The remnant polarization (P_r) and the coercive field (E_c) of PLZT 7/65/35 are of 32 μC/cm^2 and 1.2 kV/mm.

The PLZT ceramics were made by conventional solid-state reaction method. Surface of the disk-shaped ($\varphi = 20$ mm, $d = 1.5$ mm) ceramics were polished and then ultrasonically cleaned in ethanol. Copper electrodes were tightly glued on the surface of the ceramics. The emitting surface has a 0.6-mm-wide metallic electrode in a grid pattern and the rear (non-emitting) surface is covered by solid electrode (see Fig. 3). PLZT 7/65/35 samples were poled prior to emission measurements by applying a dc electric field of 3 kV/mm with the negative polarity on the grid electrode.

3. Results and discussion

3.1. Electron emission from PLZT 7/65/35 ceramics

Fig. 4 shows typical emission current waveform with a negative triggering voltage applied to the PLZT 7/65/35 ceramics. One can see that there are two distinguishable electron emission pulses per triggering voltage. The first emission pulse (pulse 1) is a relatively stable emission pulse exhibiting only 20 ns delay time. The second emission pulse (pulse 2) always occurs just after pulse 1. The position and amplitude of the two emission pulse are relatively steady. Pulse 1 has higher amplitude of 3 A and width of 200 ns than pulse 2.

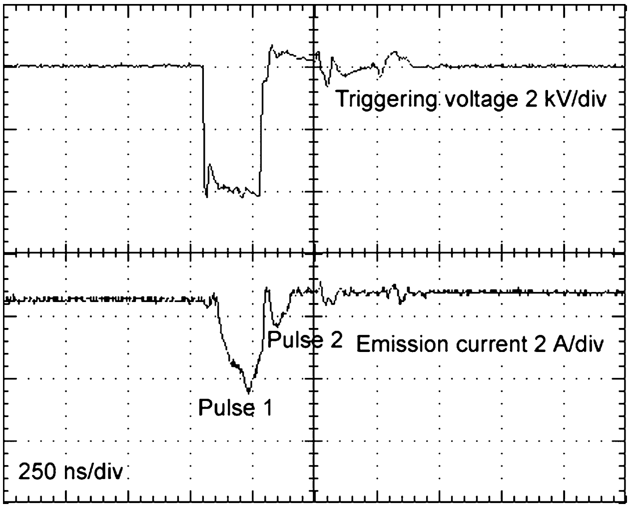

Fig. 4. Emission current waveform with a negative triggering voltage applied to the rear electrode of PLZT 7/65/35 ceramics.

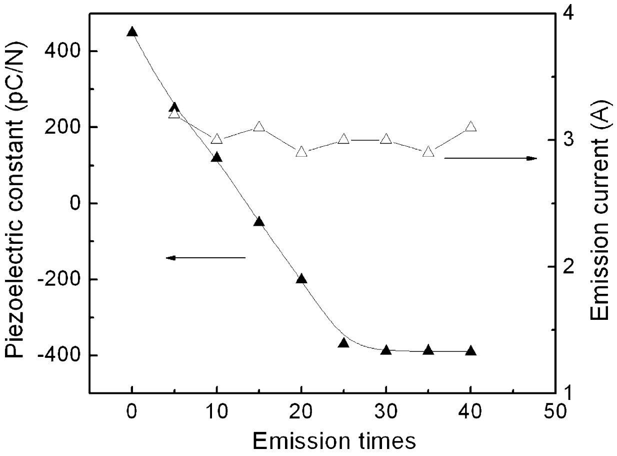

Fig. 6. Piezoelectric constant and emission current of PLZT 7/65/35 ceramics as a function of emission times (the triggering voltage was fixed to 4 kV/mm).

Fig. 5 shows electron emission current of PLZT 7/65/35 ceramics as a function of triggering voltage, indicating an increase of emission current from 0.05 to 4 A with the elevation of triggering voltage. It is thought that if the electrons are emitted by the polarization switching, there should be no emission current when the amplitude of triggering pulse below the value of coercive field (1.2 kV/mm) and the highest emission current should be obtained on condition that the triggering voltage reaches the coercive field and the increase of triggering voltage beyond the coercive field cannot promoted the emission current. However, the experiment results show that an emission current of 0.05 A was observed when the triggering voltage was 0.5 kV, which is below the coercive field and the emission current increased from 0.7 to 4 A with the promotion of the triggering voltage from 2 to 5.5 kV, which were contradicting to the explanation of the spontaneous polarization switching of the ferroelectrics.

Fig. 6 shows piezoelectric constant (d_{33}) and emission current of PLZT 7/65/35 ceramics as a function of emission times. One can see that there is a considerable decrease of piezoelectric constant (d_{33}) from 450 to −390 pC/N and a slightly fluctuation of the emission current with the increase of emission times. Because the piezoelectric constant (d_{33}) can be regarded as the representative of the order of ferroelectric domain, the emission current should diminish during the course of the decrease of the piezoelectric constant according to the explanation of spontaneous polarization switching. It was also shown that non-poled PLZT ceramics, both in ferroelectric and anti-ferroelectric phase, emit intense current when a negative triggering voltage is applied. It is difficult to explain the observed phenomena using the polarization switching model.

3.2. Electron emission from PLZT 12/65/35 ceramics

Fig. 7 shows typical emission current waveform with a negative triggering voltage applied to the PLZT 12/65/35 ceramics. The emission current waveform obtained with PLZT 12/65/35 has no significant difference with the result obtained

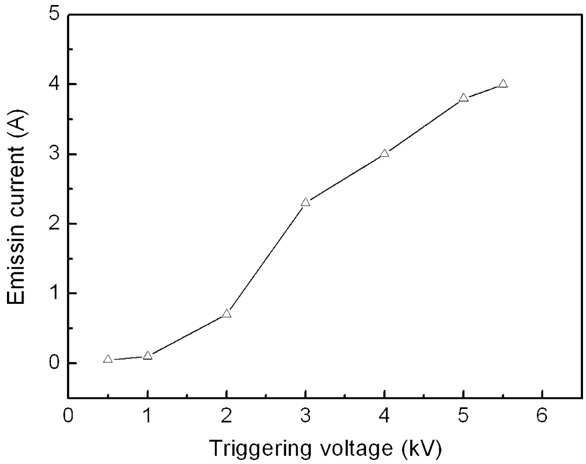

Fig. 5. Electron emission current of PLZT 7/65/35 ceramics as a function of triggering voltage.

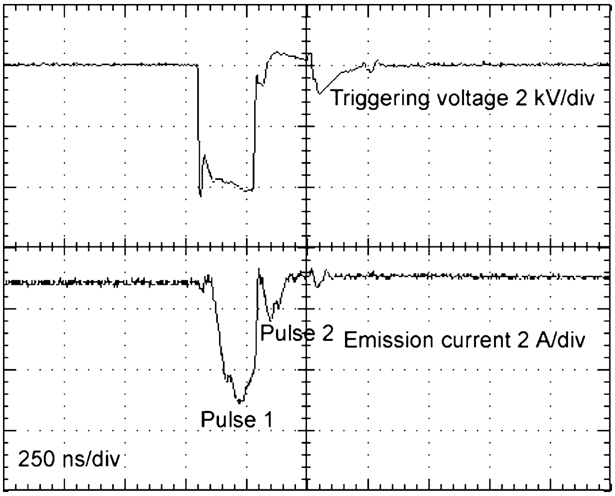

Fig. 7. Emission current waveform with a negative triggering voltage applied to the PLZT 12/65/35 ceramics.

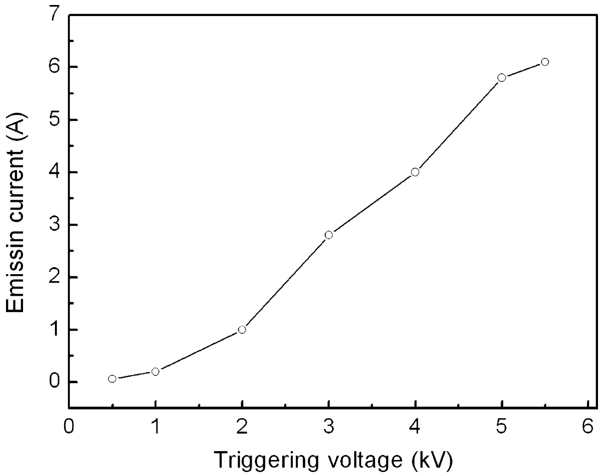

Fig. 8. Electron emission current of PLZT 12/65/35 ceramics as a function of triggering voltage.

from PLZT 7/65/35. Fig. 8 shows electron emission current of PLZT 12/65/35 ceramics as a function of triggering voltage. Comparatively, the emission current of PLZT 12/65/35 is higher than the emission current of PLZT 7/65/35, implying the PLZT 12/65/35 ceramics have a better emission property. It was shown that high-density electron emission is generated in the paraelectric phase of PLZT ceramics, which is a linear dielectric with high dielectric permittivity and only field-induced effect occurring in this material is dielectric polarization.

3.3. Mechanism of electron emission from PLZT ceramics

It was proved that spontaneous polarization does reverse when ferroelectrics subject to a fast triggering voltage. However, there is no direct evidence proved that the polarization reversal is responsible for strong electron emission phenomena. The emission characteristics together with the materials properties of PLZT ceramics led to the conclusion that strong electron emission is not caused by polarization switching. The spontaneous polarization switching is a side effect generated by the application of the high voltage pulse.

Based on the experimental results, we consider that the physical mechanism of electron emission from PLZT ceramics is field emission and surface plasma emission. The field emission is attributed to extremely high electric fields produced in the unavoidable small gaps at the metal electrode–dielectric–vacuum junction region (triple point). The enhancement of the electric field in a triple point can be roughly estimated as follow [14].

$$E = \frac{V\varepsilon}{d_0\varepsilon/d + 1} \qquad (1)$$

where V, ε and d are the trigger voltage, dielectric constant and dielectric thickness and d_0 is the micro-gap between the dielectric and metal (grid electrode). For $d_0\varepsilon/d \ll 1$, the electric field in the triple junction is approximately increased by the factor of ε. Since, the dielectric constant for PLZT 12/65/35 is about 3100, and $V = 4$ kV, $d = 1.5$ mm. So the enhancement of the electric field is 8×10^9 V/m.

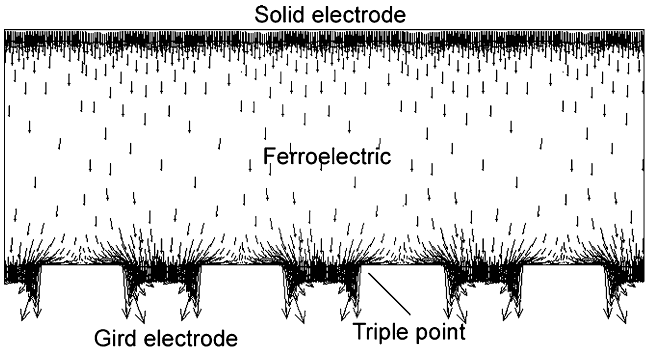

Fig. 9. Vector electric field distribution inside the ferroelectric sample.

Fig. 9 shows simulation of vector electric field distribution inside the ferroelectric sample (performed by finite element program of ANSYS 9.0). One can see that in the ferroelectric surface between the grid electrodes, the field decreases with the increase of the distance from the grid electrode. However, the highest electric field was obtained in the triple point. This high electric field has the horizontal (parallel to the emitting surface of the sample) and vertical component. The vertical component of the enhanced electric field may induce a strong bending of the bands at the boundary of triple point and vacuum allowing the electrons from the Fermi level of the ferroelectric emit into the vacuum. The classic field emission occurs, i.e., the electrons from the lattice overcome the high surface potential and emit to the vacuum. The field emission is a relative stable process and it emitted high-energy electrons from the bare surface of the ferroelectric with no apparent delay time. The characteristics of field emission are consistent with emission pulse 1. Then, the surface plasma dominates the emission process. The horizontal component of the enhanced electric field may cause acceleration of priming electrons along the ferroelectric surface with subsequently electron avalanching and surface flashover. Consequently, surface plasma generates [14]. The surface plasma will expand to the strip electrode and the bare surface of the ferroelectric. The plasma near the strip electrode has the same potential as the grounded strip electrode [15]. However, in the case of the plasma near the bare surface of the ferroelectric, a part of the plasma ions are captured by the remnant negative triggering voltage at the bare surface of the ferroelectric. The plasma may have a non-compensated negative potential. This negative potential accelerates the plasma electrons, which form the emission pulse 2. Further experiment and analysis on the surface plasma, for example, the velocity and the ingredient of the plasma, will be presented in another paper.

4. Summary

Both ferroelectric (PLZT 7/65/35) and paraelectric (PLZT 12/65/35) compositions of PLZT system were observed plasma-assisted electron emission. The emission current depends slightly on the ferroelectric properties and depends significantly on the dielectric constant. The mechanism of electron emission from PLZT ceramics was ascertained to field emission and surface plasma emission. The field emission is attributed to the high electric field enhanced by the dielectric constant. The surface plasma is induced from the triple point by the appearance of the emitted electrons together with the horizontal component of the triggering voltage.

Acknowledgement

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