

# Transparent PZT Thin Film with High Piezoelectricity on Glass Substrates

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## Abstract

*To realize high transparency and piezoelectric micro devices, Pb(Zr,Ti)O<sub>3</sub> (PZT) thin film depositions, which were conducted by Sol-Gel and Pulsed Laser Deposition (PLD) methods. This study presents the design and fabrication of multilayer thin films consisting of indium tin oxide (ITO) on glass substrates. In this study, an outstanding transmittance (85.1%) was achieved, which was fabricated by the Sol-Gel route. The Sol-Gel and PLD PZT piezoelectric devices had high inverse piezoelectric characteristics of  $|e_{31,j}|$ , 11 C/m<sup>2</sup> and 13 C/m<sup>2</sup> under 20 V<sub>Peak-to-Peak</sub> (V<sub>pp</sub>) voltage, respectively. It opens the way to low-voltage and high-piezoelectric coefficients applications for haptic devices.*

## Author Keywords

Piezoelectric, PZT thin film, Transparent, Glass

## 1. Introduction

Piezoelectric materials, especially thin films of PZT which have unique piezoelectric and inverse piezoelectric effects, are widely used in energy harvesting [1-2] or other micro-electromechanical systems (MEMS) fields [3-4]. PZT films are deposited on panels for realizing haptic feedback devices. The devices could be applied on displays, including automotive screens, smart home screens and etc [5]. Currently, for the haptic devices on displays, the high transmittance, large displacement and low driving voltage are key pivotal issues [6]. To achieve these aims, researchers are keen to develop ferroelectric films with high transmittance and piezoelectric properties. In previous studies, PZT thin films used to be fabricated on Si substrate, the PZT films with higher crystallinity and better piezoelectric response are obtained due to the better Si orientation. However, there is a growing demand for high transparent piezoelectric actuator devices with the development of haptic devices. This shows that PZT thin films on Si substrates, due to their low transmittance, face considerable restrictions when used as haptic devices. It is particularly challenging to directly deposit low-voltage, high-displacement PZT thin films onto glass substrates. Additionally, PZT thin films on ITO/Glass with a measurable piezoelectric coefficient are rarely reported because of their poor quality. To date, common techniques such as the Sol-Gel method [7], Magnetron Sputtering (RF), and Pulsed Laser Deposition (PLD) are included for PZT thin films depositions. Of these, the Sol-Gel process is commonly employed for the abilities to ensure chemical homogeneity and precise compositional control [8]. Moreover, PLD is a potential method for PZT piezoelectric thin film preparation, which can offer the advantages of high deposition rate [9]. Few studies have explored the properties of PZT piezoelectric thin films prepared via PLD method on glass

substrates. Therefore, in this work, the PZT layers will be deposited on a glass substrate using both the Sol-Gel and PLD methods, and to delve into the properties and application potential of PZT piezoelectric thin films. We deposited the 2 μm-thickness PZT layers on ITO/Glass via Sol-Gel method and PLD method. We evaluated the piezoelectric constant  $|e_{31,j}|$  by the macroscopic cantilever method, a high transverse piezoelectric coefficient  $|e_{31,j}|$  was achieved. And the entire ITO/PZT/ITO/Glass stack demonstrates a transmittance exceeding 85% in the visible spectrum and has potential as an actuator for tactile applications using the Sol-Gel approach.

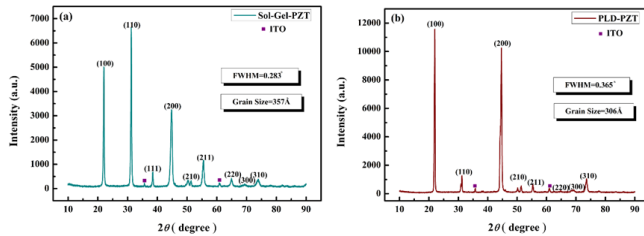
## 2. Method

We prepared the PZT/ITO/Glass samples. The high-temperature resistant glass were selected. ITO are deposited at room temperature with post-annealing at 250°C, and the thickness of 300nm/500 nm was performed by sputtering (SMD-450 BHD). The detail information about ITO preparation process can be found in literature [5]. PZT layers, with a thickness of 2 μm, were prepared on commercially sourced ITO-coated glasses via Sol-Gel and PLD methods. The detail information about Sol-Gel PZT film process also can be found in literature [5]. For PLD method, the 2-um-thick PZT films were deposited with 10% excess PbO, following a rapid thermal annealing furnace for 8 min at 650 °C, the PZT/ITO/Glass samples show better crystallization. The orientation and crystal structure of the samples on glass were measured using X-ray diffraction (D8 Discover, Bruker) with Cu-Kα radiation. Microscopes of SEM (IT500, JEOL) were used to complement the microstructural characterization including the surface and cross-section. Dielectric and ferroelectric characterization were investigated by TF Analyzer 2000 (aixACCT). The transmittance of the films on glass was assessed using a UV-Vis spectrophotometer. The piezoelectric coefficient was determined via a laser Doppler vibrometer.

## 3. Results and discussion

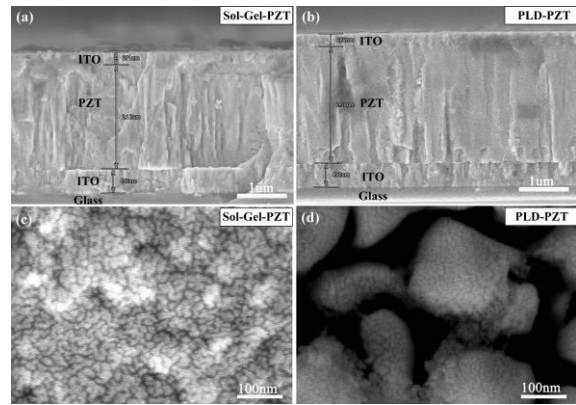
Figure 1 illustrates the XRD phase structure characteristics of PZT/ITO/Glass samples. Multiple diffraction peaks corresponding to the perovskite phase are clearly observed in the PZT/ITO/Glass samples via both Sol-Gel and PLD methods, which confirms the formation of polycrystalline PZT with a perovskite structure. Notably, the preparation method significantly influences the crystal growth of the PZT/ITO/Glass samples. For PZT/ITO/Glass samples prepared by the Sol-Gel method, the (100), (110), and (200) crystal orientations are clearly evident, with peak intensities surpassing those of random orientations, highlighting its impact on crystallographic texture. PZT/ITO/Glass samples oriented in the (100) direction show the

greatest piezoelectric properties [10]. For PLD method, the PZT/ITO/Glass samples show (100) and (110) preferred orientations, which is different from PZT/ITO/Glass samples by Sol-Gel method. It should be seen that, the PZT/ITO/Glass samples contain an additional phase, alongside the perovskite phase, which is comparable to the ITO phase structure according to the XRD PDF. And the half-peak width (FWHM), the Sol-Gel method exhibits lower performance than the PLD method, which indicates that the crystal quality of Sol-Gel method is better and the grain size is larger, this can be verified by the SEM results in Figure 2.



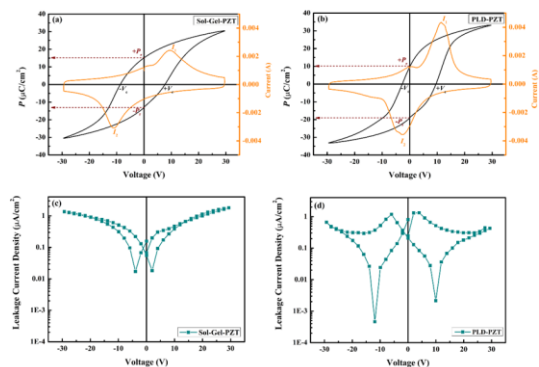
**Figure 1.** XRD pattern for the Sol-Gel and PLD PZT/ITO/Glass samples. The FWHM and grain size in the figure is calculated based on (100).

Figure 2 shows SEM micrographs of cross sections and surfaces of stacked ITO/PZT/ITO layers. SEM shows that PZT layers fabricated by Sol-Gel method display columnar structures, whereas those made by the PLD method have a smooth, compact structure. The variations in structure are due to the differing growth mechanisms of the two methods. The PZT/ITO/Glass sample is firmly attached to the substrate, and its thickness is around 2 μm. At the PZT/top ITO interface, the surface is smooth and clean, whereas the bottom ITO electrode shows a more prominent columnar structure relative to the top layer. This is likely a result of the 650 °C annealing required for wafer bonding during PZT film growth, compared to the 250 °C annealing applied to the top ITO layer following its deposition at room temperature. And the surface micrographs of the PZT/ITO/Glass samples after annealing treatment are shown in Figures 2c and 2d. Notable differences in the surface structures indicate that the preparation methods have an impact on the growth of the thin films. In Figure 2c, PZT/ITO/Glass sample deposited with the sol-gel method, this can be observed the grain boundaries are clear and the pores in the grain and grain boundaries are almost invisible, suggesting that the PZT film exhibits a relatively dense configuration. In Figure 2d, the PZT film is deposited by PLD method, and the grain size of the PZT film is not uniform, and there are obvious holes, which may affect the performance of the film. For example, an applied external voltage might concentrate at or near the defects to lead a lower voltage inside the film. This inhomogeneous voltage distribution usually corresponds to poor ferroelectric and dielectric properties [11]. The findings demonstrate that the sol-gel method produces denser PZT films compared to the PLD method, which enhances their ferroelectric and piezoelectric characteristics.



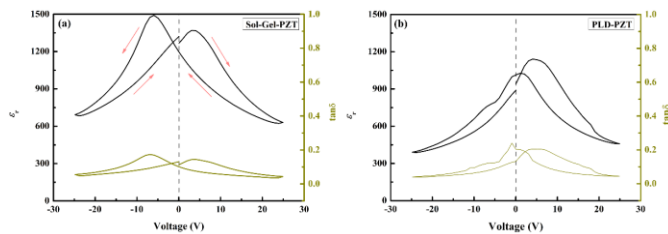
**Figure 2.** SEM images, both cross-sectional and surface views with the PZT/ITO/Glass samples.

To better evaluate the ferroelectric properties of PZT film, *P-V* hysteresis loops and *I-V* curves at 1 kHz under room temperature are shown in Figure 3a and 3b. The PZT/ITO/Glass samples was measured at 30V. Also, the obvious ferroelectric properties of the PZT/ITO/Glass samples were confirmed by observing the saturated *P-V* hysteresis loop, and the peak of the switching current (*I<sub>c</sub>*) was detected near the coercive voltage (*V<sub>c</sub>*). As seen from Figure 3a and 3b, the remanent polarization (*P<sub>r</sub>*) and *V<sub>c</sub>* of the Sol-Gel PZT/ITO/Glass sample are higher than that of the PLD PZT/ITO/Glass sample, which testifies that the Sol-Gel PZT film has higher ferroelectricity. The +*V<sub>c</sub>* and -*V<sub>c</sub>* symmetry for the Sol-Gel PZT/ITO/Glass sample is higher, which testifies that the Sol-Gel PZT/ITO/Glass sample has better crystallization properties, and also introduces fewer defects correlated with the microstructure characteristics shown in Figure 2. To better prove the occurrence of defects, leakage current experiments were conducted, and the results are illustrated in the Figure 3c and 3d. In the application of piezoelectric devices, leakage current of PZT/ITO/Glass samples is a very key issue. While ferroelectric films are made into a device, leakage current will inevitably occur under an applied voltage. Low leakage current is the goal of the prepared device. It can be observed that as the voltage increases, the leakage current density slowly increases until it reaches saturation, but the area ratio of the leakage current curve of PLD PZT/ITO/Glass sample is larger than sol-gel PZT/ITO/Glass sample, which indicates that PLD PZT/ITO/Glass sample has more current reactive power loss, which confirms the existence of defects in PLD preparation. It shows that, in this work, the PLD PZT/ITO/Glass sample is not suitable for the application of high ferroelectric properties devices.



**Figure 3.**  $P$ - $V$  hysteresis loops,  $I$ - $V$  curves, leakage current characteristic measured at room temperatures for Sol-Gel and PZT/ITO/Glass samples.

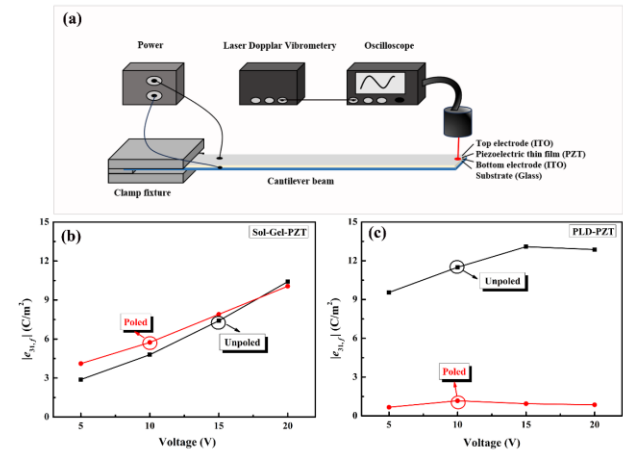
This work also studies the change loops of the dielectric permittivity -bias electric voltage ( $\epsilon_r$ - $V$ ) and dielectric loss-bias electric voltage ( $\tan\delta$ - $V$ ) of Sol-Gel and PLD PZT/ITO/Glass samples. The  $\epsilon_r$ - $V$  relationship reflects the switch in domain structure with DC bias voltage. From Figure 4, PZT/ITO/Glass samples show a typical nonlinear butterfly-like curve under a DC bias voltage close to 25 V, proving the characteristics of good ferroelectric and piezoelectric properties. Figure 4a gives that  $\epsilon_r$  is 1500, this is larger than the value for PZT/ITO/Glass samples deposited via the Sol-Gel method reported in the literature [6]. Besides, both the  $\epsilon_r$ - $V$  curves in Figure 4a and 4b are off-center, which is caused by the internal bias voltage of the films. It is reported that the internal bias voltage is caused by the existence of defects [12]. The permittivity of PLD PZT/ITO/Glass samples is reduced compared to that of Sol-Gel PZT/ITO/Glass samples. This shows that the PLD PZT/ITO/Glass sample has many defects, which leads to a large internal bias voltage and obvious pinning effect that affects domain switching, this can be verified by its higher dielectric loss.



**Figure 4.** Dielectric properties for the PZT/ITO/Glass samples.

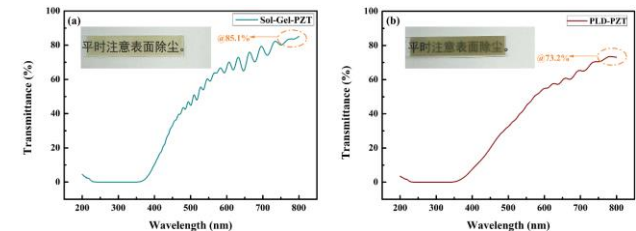
The transverse piezoelectric coefficient ( $|e_{31, j}|$ ) is the most important factor to be considered for haptic device applications. To evaluate the piezoelectric properties, the device with an area of  $15 \times 3 \text{ mm}^2$  is mounted on a measurement system. A setup schematic is illustrated in Figure 5a. The computed inverse piezoelectric coefficient versus the applied voltage  $V$  is shown in Figures 5b and 5c. The PZT/ITO/Glass samples exhibit notable piezoelectric properties, and the transverse piezoelectric coefficients is approximately  $10 \text{ C/m}^2$ , comparable to typical PZT films on silicon substrates and superior to those on metal substrates [13-16]. The PZT/ITO/Glass samples were subjected to a  $10 \text{ V}_{DC}$  polarization to evaluate the effect of DC polarization on their piezoelectric properties. For unpoled Sol-Gel PZT/ITO/Glass sample, the converse piezoelectric coefficient increases gradually with applied voltage and the  $|e_{31, j}|$  is at  $3 \text{ C/m}^2$ - $11 \text{ C/m}^2$  from  $5 \text{ V}_{pp}$  to  $20 \text{ V}_{pp}$ . After poling, the converse piezoelectric coefficient at low voltage increases obviously, linearity of  $|e_{31, j}|$  is attributed to the extrinsic contributions due to  $180^\circ$  domains switching [17] and the polarization is conducive to the complete switch of the domain [18, 19]. However, for poled PLD PZT/ITO/Glass samples, the general piezoelectric coefficient decreases greatly due to the large number of defects caused the pinning effect to hinder the switching of domains. From the point of view of piezoelectric applications, the piezoelectric layer of Sol-Gel PZT/ITO/Glass sample can be

determined by the final properties of the device due to its better piezoelectric coefficient. Indeed, stable and high piezoelectric properties are very necessary for device applications.



**Figure 5.** (a) A schematic of the measurement setup for the displacement response. (b)(c) Piezoelectric coefficient as a function of applied voltage for the PZT/ITO/Glass samples.

With the advent of haptic devices on glass touch screens and smart Windows. There is a growing demand for transparent piezoelectric transducers. The optical transmittance of Sol-Gel and PLD PZT/ITO/Glass samples was tested using the spectrophotometer, the wavelength range is 200 to 800nm. The results are exhibited in Figure 6, the visible light wavelength range is 200 to 400 nm, the transmittance of Sol-Gel PZT/ITO/Glass sample is equivalent to that of PLD PZT/ITO/Glass sample in this range, and the transmittance value is about 0%. What's more, The Sol-Gel PZT/ITO/Glass sample exhibits a slightly higher transmittance than the PLD PZT film in the 400 to 800 nm wavelength range, likely due to the porosity present in the film, as shown in Figure 2, which is up to 85.1%, which is comparable to the transmittance of commercial ITO glass substrate (84%) [20]. Furthermore, the generation of oscillatory behavior is observed in both PZT/ITO/Glass samples, the phenomenon is explained by the existence of multiple interfaces in the PZT layer, leading to reflections that cause interference among the transmitted beams. The emergence of this oscillation characteristic also proves that the prepared film has a smooth surface and uniform thickness. Due to the high optical transmittance of Sol-Gel PZT/ITO/Glass sample, the underlying text feature is clearly visible (Figure 6a), indicating that it has broad application prospects in the haptic field.



**Figure 6.** Optical transmittance characteristics for the PZT/ITO/Glass samples. Insert shows optical photographs.

In conclusion, we successfully deposited a 2  $\mu\text{m}$  polycrystalline PZT film on an ITO-coated glass substrate using both the Sol-gel method and PLD method. A functional ITO/PZT/ITO capacitor with notable piezoelectric properties was fabricated on the glass substrate. The PZT/ITO/Glass samples prepared via the Sol-Gel method and PLD method exhibit a pronounced inverse piezoelectric effect, with  $|e_{31,f}|$  values of 11 and 13  $\text{C}/\text{m}^2$  at a driving voltage of 20  $\text{V}_{\text{pp}}$ , respectively. The entire stack demonstrates a transmittance exceeding 85.1% in the visible spectrum and has potential as an actuator for tactile applications using the Sol-Gel approach. These findings indicate that nearly transparent piezoelectric film devices can be realized through the Sol-Gel method, paving the way for expanded applications in the field of tactile devices.

#### 4. Acknowledgements

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