Measurement of piezoelectric coefficients of lead zirconate titanate thin films by strain-monitoring pneumatic loading method

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A method to simultaneously measure the longitudinal ($d_{33}$) and transverse ($d_{31}$) piezoelectric coefficients of a lead zirconate titanate (PZT) thin film was developed. This system was based on the pneumatic loading method but was modified to monitor the radial strain when a pressurized gas was introduced into the chamber. The results of the bulk piezoelectric material measured by this system coincided with that measured by both the Berlincourt method and the resonance method. The effective $d_{33}$ and the real $d_{31}$ of the PZT thin film fabricated by the sol-gel multiple coating method, and poled at 300 kV/cm were 125 and $-60$ pC/N, respectively. The real $d_{33}$ estimated upon considering the constraints by the silicon substrate was 180 pC/N. © 2002 American Institute of Physics. [DOI: 10.1063/1.1487901]

Lead zirconate titanate [Pb(Zr,Ti)O$_3$] (PZT) thin films have been studied extensively for their applications not only in memory devices, especially, nonvolatile memory (FRAM), but also in microelectromechanical system applications. Piezoelectric devices such as sensors and actuators, including micromotors and micropumps, can be fabricated at low cost using silicon technology. However, it is necessary to know the longitudinal ($d_{33}$) and transverse ($d_{31}$) piezoelectric coefficients of the film precisely in order to design microsensors and actuators. Due to the constraints imposed by the substrate, the piezoelectric coefficients of a thin film cannot be measured directly using the standard resonance method. Therefore, a static or quasistatic method needs to be used. There are two trends in the evaluation technique for piezoelectric films. One is to use the direct piezoelectric effect such as the normal loading method, impulse method, and wafer flexure technique; the other is to use the converse piezoelectric effect such as the interferometer method and the atomic force microscopy method.

The laser interferometer method is a well-established method for the characterization of both longitudinal and transverse piezoelectric coefficients of piezoelectric thin films. It is widely accepted that reasonable effective $d_{33}$ values are obtained by a double beam interferometer. However, this method requires precise optical alignment and a meticulous operation. Furthermore, in the case of the $d_{31}$ measurement, appropriate sample preparation (cantilever beam) is necessary. The pneumatic loading method is an alternative method for characterizing the longitudinal piezoelectric coefficients. This method has an advantage over the normal loading method because this generates a uniform uniaxial stress using a pressurized gas as a loading medium. However, the results measured by this method always contain the effect of the in-plane stress caused by the O-ring movements as the chamber is pressurized. The strain-monitoring pneumatic loading method (SMPLM) described in this letter is a method for characterizing the piezoelectric coefficient accurately. Moreover, it is capable of measuring longitudinal ($d_{33}$) and transverse ($d_{31}$) piezoelectric coefficients simultaneously with a simple sample preparation and measuring operation.

The experimental setup schematically shown in Fig. 1 is basically same as that suggested by Xu et al.; the only modification is the strain gauge attached on the bottom of the substrate. The pressure applied in this experiment ranged from 0 to 0.7 MPa, and was monitored using an Omega PX602 pressure transducer. The induced charge was collected using a Kistler 5011B charge amplifier, and the radial strain was measured using a TML FLA-1-11 strain gauge and a TML DA-16A strain gauge amplifier. The voltage outputs from the charge amplifier and the strain gauge amplifier were monitored in real time by a computer.

In order to calibrate the system, the piezoelectric constants of a bulk PZT were measured and compared to those measured by the quasistatic piezoelectric $d_{33}$ meter (ZJ-3D, Institute of Acoustics, Academic Sinica, Beijing, China) and the standard resonance method. A commercially available

![FIG. 1. Schematic drawing of the experimental setup for the SMPLM.](image)

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PZT (APC-841, APC International Ltd., PA) was machined into 25 mm by 25 mm by 1 mm in size and subsequently polished on both sides with a 6 μm diamond paste. The electrodes were formed by a silver paste and were annealed at 600 °C for 30 min. The bottom electrode was formed on one entire face, but the top electrode was 5 mm in diameter. The specimens were poled by applying 30 kV/cm at 120 °C for 10 min. The Young’s modulus and Poisson’s ratio of the bulk specimen were measured using the pulsed echo overlap method.

The charge and the strain of the PZT specimen according to the applied pneumatic pressure are shown in Fig. 2. When the gas pressure was applied to the sample, the induced charge increased suddenly to a maximum, then decreased slowly until it is stabilized after approximately 90 s. On the other hand, the radial strain increased slowly after a sudden jump until it reached a steady state value. This is believed to be due to a viscoelastic behavior of the O rings holding the specimen. In other words, when the chamber is pressurized rapidly with the gas, the O rings expand outward instantly at first and then slowly until they reach an equilibrium state. Therefore, the induced charge decreases gradually (Fig. 2) as the radial tensile stress on the specimen increases slowly.

The electrical displacement and the radial strain of the specimen as a function of pressure are shown in Fig. 3. The electrical displacement curve at the constant strain region in Fig. 3 corresponds to \( d_{33} \) and the \( y \)-axis intercept is related to \( d_{33} \). The \( d_{33} \) and \( d_{31} \) of the specimen estimated by this method were 311 and \(-70\) pC/N, respectively. When the same specimen was examined by using the conventional methods (Berlincourt meter and the resonance method), the \( d_{33} \) and \( d_{31} \) were 304 and \(-70\) pC/N, respectively. This excellent agreement supports the validity of the present SMLM method.

This method was then applied to PZT thin film. The PZT film was fabricated on commercially available Pt(111)/Ti/SiO\(_2\)/Si (Inostek Inc., Seoul, Korea) substrates by using a spin coater (Laurell Tech. Inc., North Wales, PA). The 1-μm-thick crack-free PZT film was fabricated by a multiple coating method. The composition of the film was Pb(Zr\(_{0.52}\)Ti\(_{0.48}\))O\(_3\), which is the morphotropic phase boundary,\(^{13}\) and the Young’s modulus of the wafer was 150 GPa.\(^{14}\) The top electrode was formed by sputtering a thin Pt film through shadow mask with an array of 2.5 mm diameter holes. It is defined as a being in the positive poling state when a positive voltage is applied to the top electrode. Poling was performed at room temperature for 100 s.

Figure 4 shows the piezoelectric hysteresis loop of the 1-μm-thick PZT thin film with 52/48 composition. The piezoelectric coefficients were saturated at approximately 300 kV/cm, and the measured \( d_{33} \) and \( d_{31} \) were 125 and \(-60\) pC/N, respectively. These values match very well with previous results.\(^{9,12}\) While the \( d_{31} \) measured by this method reflects the real value, the measured \( d_{33} \) is not the actual \( d_{33} \) but an effective \( d_{33} \) due to the substrate constraints. Lefki
and Dormans suggested the relationship between the effective $d_{33}$ and the real $d_{33}$. The relationship was estimated by Xu et al. as follows:

$$d_{33}(dp) = d_{33} - \frac{2d_{31}(s_{11}^{E} + \nu Y)}{s_{11}^{E} + s_{12}^{E}} = d_{33} + 0.96d_{31}, \quad (3)$$

where $d_{33}(dp)$ represents the effective $d_{33}$ measured by the direct piezoelectric effect, $s_{ij}^{E}$ and $d_{ij}$ are the compliances and piezoelectric coefficients of the film, and $Y$ and $\nu$ are the Young’s modulus and the Poisson’s ratio of the substrate. The real $d_{33}$, calculated using the published data, was 180 pC/N at 300 kV/cm.

In summary, the SMPLM is a simple procedure for accurately and simultaneously measuring the piezoelectric coefficients of thin films. The effective $d_{33}$, the real $d_{33}$, and the real $d_{31}$ of the 1-μm-thick PZT thin film with 52/48 composition poled at 300 kV/cm for 100 s were 125, 180, and −60 pC/N, respectively.