Longitudinal piezoelectric coefficient measurement for bulk ceramics and thin films using pneumatic pressure rig

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A pneumatic pressure rig was designed to measure the effective d_{33} coefficient of thin film piezoelectrics by applying a known stress and monitoring the induced charge. It was found that the stress state imposed included components both perpendicular and parallel to the film plane. The later were due to friction and could largely be relieved through sliding of the O-rings to their equilibrium positions for a given pressure. The induced charge stabilized as equilibrium was reached and most of it was produced by the normal component of the stress. By minimizing the surface friction and compensating for the remnant in-plane stress, very good agreement was obtained among the d_{33} values measured by the Berlincourt method, double-beam interferometry and this method for a bulk lead zirconate titanate (PZT) sample. The d_{33} value of PZT thin films made by sol-gel processing was also measured. The as deposited films usually showed very weak piezoelectricity with d_{33} values ranging from 0 to 10 pC/N, indicating little pre-existing alignment of the domains. With increasing poling field, the d_{33} value also increased and saturated at poling fields exceeding three times the coercive field. Typically, films with thicknesses around 1 μm had d_{33} values of 100 pC/N. Good agreement between double-beam interferometry and this technique was also obtained for thin films. The small difference between the two measurements is attributed to the effect of mechanical boundary conditions on the effective d_{33} coefficient. *© 1999 American Institute of Physics.* [S0021-8979(99)05813-2]

I. INTRODUCTION

There has been a great deal of interest paid to the design and fabrication of microelectromechanical systems (MEMS) in recent years. Micromechanical actuators based on piezoelectricity are particularly attractive due to the high energy densities, and high-transmitted forces and torques which can be achieved.[1,2] Piezoelectric devices such as sensors and actuators can be fabricated at low cost using silicon technology designed for integrated circuits. A variety of applications for piezoelectric thin films integrated on silicon substrates, including micromotors,[2,3] accelerometers,[4] and microsonar arrays,[5] have been described.

For the design of miniature sensors and actuators, it is desirable to have a comprehensive knowledge of material properties such as the elastic and piezoelectric constants. However, for piezoelectric thin films, reliable measurements of the piezoelectric coefficients are still not widely available. Due to the constraints imposed by the substrate, the piezoelectric coefficients of thin films cannot be directly measured through standard resonance methods,[6] thus static or quasistatic methods have to be used. These methods can be categorized by whether the direct piezoelectric effect, i.e., applying a stress and measuring the induced charge, or the converse piezoelectric effect, i.e., applying a voltage and measuring the induced elongation, is used. It should be pointed out that in these measurements, the ratio between charge and stress (or strain and voltage) does not represent the piezoelectric coefficient of the free sample, but an effective coefficient since the film is clamped to the substrate.

For converse measurements of the effective d_{33}, the longitudinal elongation of the film is on the order of 0.1–100 Å due to the small film thickness ($\sim\mu$m). Laser interferometry has been used to measure these displacements.[7,8] Although reasonable values of effective d_{33} have been reported for both single-beam and double-beam interferometer systems,[9,10] reliable measurement using a single-beam instrument is very difficult because of both bending and backside motion of the sample.[10,11] Double-beam interferometry is able to measure the film dilatation accurately by eliminating the influence of substrate motion (including bending), but it requires a more sophisticated optical system and the measurement is strongly influenced by the optical alignment.

On the other hand, although the direct piezoelectric effect has been widely used in determining d_{33} for bulk piezoelectric materials, little success has been achieved for piezoelectric thin films. The major obstacle which prevents this kind of measurement for thin films is believed to be the simultaneous bending of the sample when force is applied perpendicular to the film plane to produce a uniaxial stress. In a thin film sample, a very thin layer of piezoelectric material is rigidly clamped to a much thicker substrate. As a result, even a small substrate bending can generate very large biaxial stresses in the piezoelectric thin film, which in turn produces a large amount of electrical charge through the transverse piezoelectric effect. This makes the accurate measurement of the charge induced by the applied uniaxial stress very difficult. Another problem for thin film d_{33} measurements is that the small thickness of the sample makes stress alignment very difficult.

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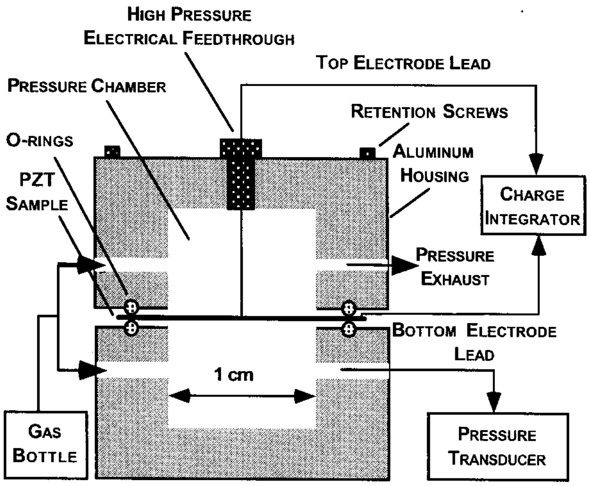


FIG. 1. Schematic drawing of the experimental setup for d_{33} measurement.

In this article, a pneumatic pressure rig designed to apply a uniaxial stress to piezoelectric thin films is described. The piezoelectric charge response of both bulk and thin film lead zirconate titanate (PZT) samples due to the applied stress was studied over the pressure range from 0 to 1.2 MPa. It was found that a component of in-plane stress led to inflated d_{33} values in both bulk and thin film samples. A measurement procedure which eliminated the error by self-compensating the remnant in-plane stress was then developed.

II. EXPERIMENTAL PROCEDURE

The experimental setup developed in this work is shown schematically in Fig. 1. There were two major parts of this setup: a pneumatic pressure rig which was designed to apply a uniform uniaxial stress to the tested sample and a charge integrator. The stress rig consisted of two identical aluminum components machined with cavities 1 cm in diameter. The sample was placed between the aluminum fixtures with cavities both above and beneath it. The two cavities were connected so that the pressure inside them was always equal. O-rings were used on both sides of the sample for sealing. To reduce the surface friction, Parker O-lube lubricant was applied between the O-ring and the sample. By introducing high-pressure nitrogen gas into these cavities simultaneously, forces were imposed which acted uniformly and equally on both sides of the sample, thus a uniform uniaxial compressive stress was applied to it. Similarly, releasing the pneumatic pressure resulted in a uniaxial stress of opposite sign. The pressure inside the two cavities was measured using Omega PX602 pressure transducers. The pressure applied in this experiment was in the range from 0 to 1.2 MPa. Because the applied force was balanced everywhere across the faces of the sample, the application of pneumatic pressure would not bend the sample even if it had an initial curvature due to the thermal expansion coefficient mismatch between the piezoelectric film and the substrate. Another advantage of using pneumatic pressure is that a uniaxial stress can be obtained without the need to align the stress rig, even though the film thickness is much smaller than the lateral dimension of the sample. Unlike the normal load method, because there is no solid contact between the sample surface and the gas, uniform stress can be produced easily on a sample with nonflat surfaces (which is usually the case for piezoelectric thin films due to the finite thickness of the top electrode).

The induced charge produced upon applying or releasing pressurized gas was collected using a charge integrator[12] which converted the collected charge into a variation of voltage on a capacitor of known size placed in series with the stressed sample. Since the circuit was in virtual ground mode, the voltage between the two electrodes of the sample was always zero, so that almost all the induced charge was driven to the capacitor. The voltage output from the charge integrator was monitored in real time using a Hewlett Packard 54600A oscilloscope.

The piezoelectric coefficient d_{33} (bulk material) or effective d_{33} (thin film) was calculated using the following equation:

$$d_{33}=\partial D_3/\partial T_3=\Delta Q/(\Delta P^*A), \qquad (1)$$

where D_3 and T_3 are the electrical displacement and mechanical stress in thickness direction, ΔP is the change of the cavity pressure, ΔQ is the measured electric charge induced by that pressure change, and A is the area of the electrode.

The charge response of both a PZT bulk ceramic sample and PZT thin films were investigated in this work. The bulk ceramic studied was a piece of PZT-5A which was 25 mm by 25 mm by 2 mm in size and polished with 1 μm alumina powder on both sides. A thin layer of Pt was sputtered on one entire face for the bottom electrode. On the other side, a 7.5 mm diameter top electrode was also formed with sputtered Pt. The sample was then poled at 100 °C for 10 min under an electric field of 20 kV/cm. PZT thin films were prepared by the sol-gel method. The procedure for the preparation of the sol-gel films can be found elsewhere.[13] The substrate used was platinum-coated, oxidized silicon wafer with a thin titanium adhesion layer. The composition of the film was $Pb(Ti_{0.48}Zr_{0.52})O_3$, which is at the morphotropic phase boundary. Top electrodes were formed by sputtering a thin platinum layer through a shadow mask with an array of 1.6 mm diameter holes. The back of the sample was polished with 1 μm alumina powder. Air dry silver epoxy was used to contact the top and bottom electrodes of the thin film capacitor. Poling was performed at room temperature.

The double-beam interferometer measurements of d_{33} for both bulk and thin film PZT samples were made using a system described elsewhere.[7,8] A ZJ-2 piezoelectric d_{33} meter (Institute of Acoustics, Academia Sinica) was also used to measure the d_{33} value of the bulk PZT sample. The dielectric constant and loss factor were measured using an HP4192 LCR meter, and the ferroelectric hysteresis loop was measured using a RT66A ferroelectrics tester (Radiant Technologies).

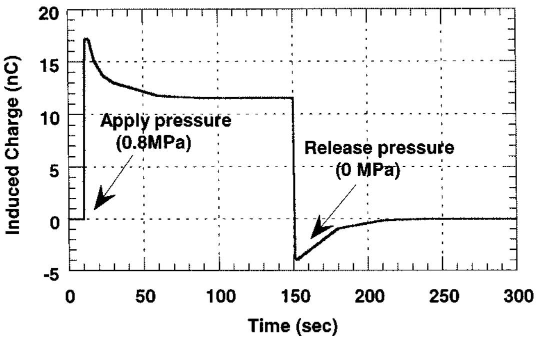


FIG. 2. Charge response of the bulk PZT sample to applied pneumatic pressure.

III. RESULTS AND DISCUSSION

A. Measurements on bulk PZT

1. Charge response and stress analysis

The charge response of the bulk PZT sample upon applying and releasing pressure was investigated first to study the stress state induced by pneumatic pressure. As shown in Fig. 2, when pressurized gas was introduced into the rig, the induced charge quickly reached a maximum, followed by a continuous asymptotic decrease, until it stabilized after about 90 s. The measured pressure in both cavities was equal and constant throughout the signal stabilization. The signal measured after releasing pressure was very similar to the signal on applying pressure, and the final values of the signal at stabilization were almost equal in magnitude, though opposite in sign. It should be emphasized here that the variation of the measured induced charge was not due to the leakage either from the PZT sample or the electronics. This was verified by applying a constant normal load to this sample. A step signal was indeed obtained, as expected. This result indicated that the time dependence of the induced charge observed on the oscilloscope was caused by some kind of change in the stress applied to the PZT sample. Since the pressure was constant inside the cavities, there was no change in the stress which was perpendicular to the sample plane. Obviously the pneumatic pressure also generated an additional stress component, and this stress was time dependent.

We have demonstrated that the in-plane stress induced by friction between the O-ring and the sample surface was the origin of the above observation.[14] The friction arises when there is a pressure change introduced into the cavities. Increasing the pressure increases the force acting on the O-ring parallel to the surface and pushes the O-rings outwards. This force generates surface friction on both sides of the sample, so that an equivalent tensile in-plane stress is created. The in-plane stress decreases over time as the pneumatic pressure acting on the O-ring leads to O-ring sliding. O-ring sliding reduces the net force acting on the O-ring by creating a restoring force via deformation. Eventually the in-plane stress stabilizes as a force balance on the O-ring is reached. The net result is that in addition to the desired compressive stress normal to the wafer surface created by pressurizing the cavities, remnant tensile in-plane stress is also produced by the pneumatic pressure at the steady state. For the same reason, a compressive in-plane stress arises when pressure is released from the cavity, since the unbalanced O-ring restoring forces cause the O-rings to slide back towards their original positions. Since d_{31} and d_{33} have opposite signs in PZT, the piezoelectric charges induced by the normal component of the stress and the in-plane stresses add to each other. With the decrease in the in-plane stress over time, the total amount of induced charge also decreases. Finally, at steady state, both stresses are constant and the signal no longer changes with time. At this point, the in-plane stress was at its minimum since a majority of it had been released through O-ring deformation or recovery. Since the induced charge measured at this stage contains the smallest signal induced by the in-plane stress component, it was used to approximate the d_{33} coefficient using Eq. (1).

2. Remnant in-plane stress analysis

To evaluate the remnant in-plane stress level at stabilization and the error in the calculated d_{33} introduced by that stress, a calibration was done by comparing the calculated d_{33} values with values measured by other techniques. A bulk PZT ceramic sample measured using the Berlincourt method and double beam interferometry gave d_{33} values of 305 and 302 pC/N, respectively, indicating a very good agreement between these two techniques. However, the calculated d_{33} from the pneumatic pressure method was always larger than those values, showing an appreciable amount of remnant in-plane stress present at stabilization. Depending on the experimental conditions which controlled the in-plane stress, the calculated d_{33} values were about 3%–10% larger than those measured by the other two techniques. To eliminate this error, a detailed investigation was made on both the factors which controlled the magnitude of the remnant in-plane stress as well as how to reduce it by optimizing the design of the pressure rig and the measurement procedure.

Since the frictional force is given by $F=\mu N$, where μ is the friction coefficient between the two surfaces and N is the force parallel to the surface, any factor which influences μ or N affects the friction. In our experiments, the following factors were found to be particularly important in friction control: the O-ring compression (i.e., the percentage reduction in dimension in the clamping direction), lubrication, sample surface roughness, the O-ring cross-sectional area and the O-ring hardness. Figure 3 shows how the measured pressure-induced charge was affected by O-ring compression. The results indicated that the remnant in-plane stress dropped with decreasing O-ring compression. This was due to a smaller μ when the O-ring was less clamped. Therefore very small O-ring compression was favorable in this application, especially since a little leakage of gas from the cavities is permitted. Since rubber has an inherently high friction coefficient with almost all metallic and nonmetallic surfaces, adequate lubrication is very important, especially for pneumatic seal applications. A good lubricant can form a strong film on the sample surface which the O-ring cannot wipe away, thus reducing the friction coefficient. It was found in our experiment that the friction was greatly reduced by the application of lubricant, and reliable measurements were ob-

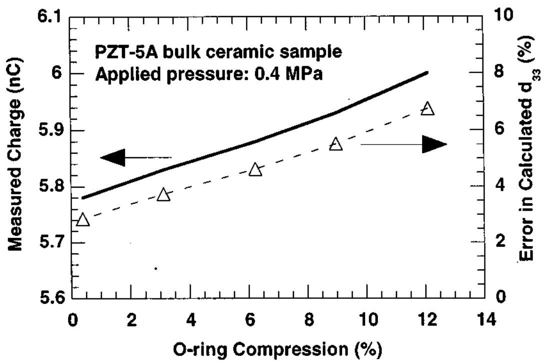
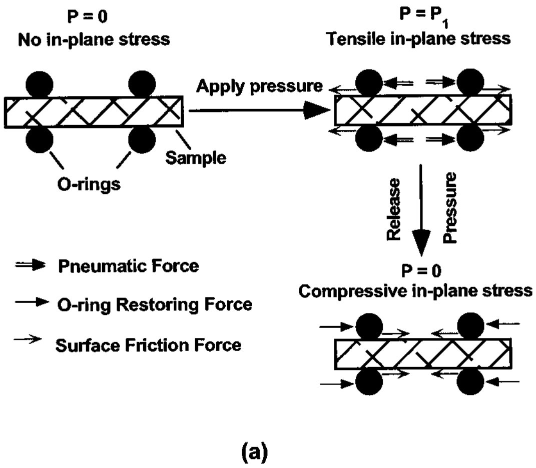


FIG. 3. Effect of O-ring compression on measured pressure-induced charge. O-ring compression was calculated as $(d_0 - d_c)/d_0$, where d_0 is its original diameter and d_c is the dimension in the clamped direction. The error in the calculated d_{33}, was estimated by comparing the calculated d_{33} value with that measured by Berlincourt meter.



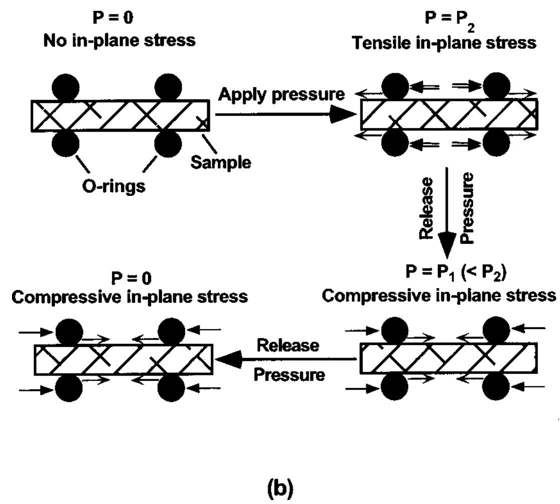


FIG. 4. Remnant in-plane stress produced by pneumatic pressure during (a) procedure in which pressure was switched between 0 and P_1, (b) procedure with a remnant in-plane stress self-compensation mechanism.

tained only when the sample was properly lubricated. Friction was also reduced by polishing the sample surfaces using 1 μm alumina powder. The smoother the surface, the smaller the remanent in-plane stress. Smaller remnant in-plane stresses were also obtained when an O-ring with a smaller cross-sectional area was used, since this reduced the pneumatic force acted on it. The reduction of friction through use of a harder O-ring is due to its smaller friction coefficient.

Under the best experimental conditions used to reduce the in-plane stress component, the d_{33} value calculated from the measured pressure-induced charge exceeded that measured by Berlincourt meter and interferometry by about 3%–4%. This suggests that the error in a d_{33} coefficient derived from this measurement would be under 5%. However, this optimized condition was not always readily achievable. As a result, there was a relatively larger scatter in the derived d_{33} value, and sometimes errors as large as 10%–15% were observed.

3. Self-compensation of remnant in-plane stress

To further improve the accuracy of the d_{33} measurement and more importantly, the reliability of this charge-pressure measurement technique, a measurement procedure for remnant in-plane stress self-compensation (RIPSSC) was developed. In this method, the charge contribution from the in-plane stress induced charge could be totally eliminated by manipulating the remnant stress levels. Figure 4 shows schematically how this method works.

In the previous section, where the induced charge was measured during switching the pressure between $P=0$ and $P=P_1$ [Fig. 4(a)], the remnant in-plane stress at the two steady states were of opposite sign. The change in in-plane stress between the two measurement states is fundamentally responsible for the inflated d_{33} values. The idea of the RIPSSC method is to tailor the remnant in-plane stress so that they are of the same sign and magnitude in the two steady states. If this is achieved, then there will be no change in in-plane stress between the two steady states. The induced charge is then only that due to the normal component of the stress, and accurate d_{33} values can be obtained. One way to reach this condition is to preload the cavities with a pressure $P=P_2$ which is higher than P_1 and then reduce the pressure to $P=P_1$ [Fig. 4(b)]. By doing so, a compressive remnant in-plane stress was imposed on the sample due to the O-ring recovery. After stabilization, the pressure is then released to $P=0$ and the induced charge is measured between the steady states at $P=P_1$ and $P=0$.

The dependence of the remnant in-plane stress at P_1 on the preloading level P_2 was investigated for the purpose of controlling the magnitude of the remnant in-plane stress. Figure 5 shows that the induced charge measured between $P=P_1$ and $P=0$ decreased with an increase in the preloading pressure (i.e., as P_2 increased). If Q' is the charge which is due to the normal component of the stress (calculated using the d_{33} value obtained from the Berlincourt measurement), then the measured charge Q was equal to Q' when the preloading pressure P_2 was about two times the value of P_1. This indicates that the remnant in-plane stress at P_1 can be

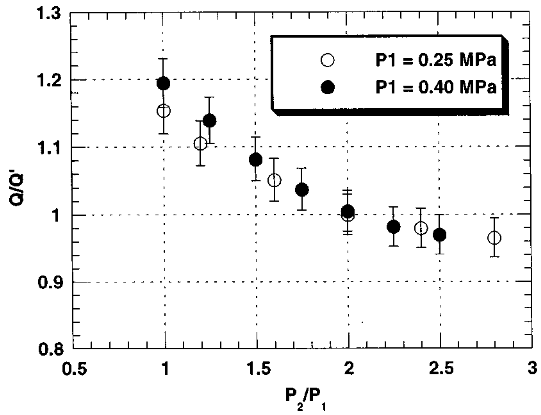


FIG. 5. Influence of the preloading pressure P_2 on the measured charge for a pressure change from $P=P_1$ to $P=0$. Q is the measured pressure-induced charge, and Q' is the charge due to the normal component of the stress evaluated using the d_{33} value measured by Berlincourt meter.

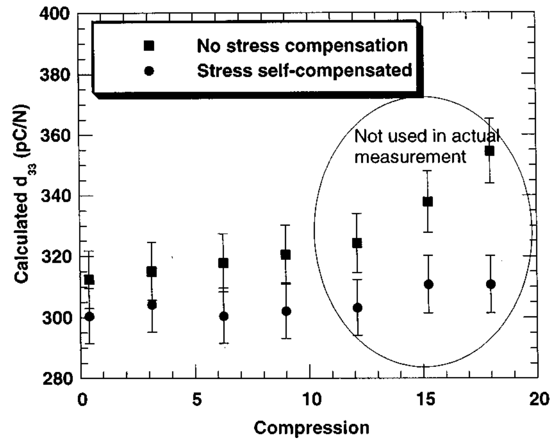


FIG. 7. Influence of O-ring compression on the measure d_{33} coefficient of bulk PZT-5A. The effectiveness of stress compensation is clear.

controlled by the preloading pressure P_2, and the remnant in-plane stress level is controlled mainly by the pressure change between the two steady states. This is reasonable since the friction is proportional to the pressure change. Thus, one might expect that the remnant in-plane stress due to the friction would also be proportional to the change of pressure.

Using the RIPSSC method, a linear relationship between the induced charge and the applied pressure was obtained (see Fig. 6). d_{33} for the bulk PZT-5A ceramic sample derived from the slope of the curve was 304 pC/N, which was in very good agreement with the value obtained by both the Berlincourt method and interferometry, indicating that the influence of surface friction on the d_{33} measurement was eliminated by the RIPSSC method.

In addition to enhancing the measurement accuracy, the RIPSSC method also made the d_{33} measurement much less sensitive to variations in the remnant in-plane stress. Figure 7 shows the influence of the O-ring compression on the measurement results with and without remnant in-plane stress self compensation. In this experiment, very large compressions (which were not used in actual d_{33} measurements) were intentionally applied to the O-rings. As the O-ring compression increased, friction increased and so did the remnant in-plane stress, as indicated by a considerable increase (15%) in the calculated d_{33} value when the RIPSSC method was not employed. On the other hand, there was only a 2% variation in the measured d_{33} value from zero compression up to 17% compression when the RIPSSC method was used. Practically, this is very important in the routine application of the technique, making the measurements much more consistent and the measurement procedure much more convenient.

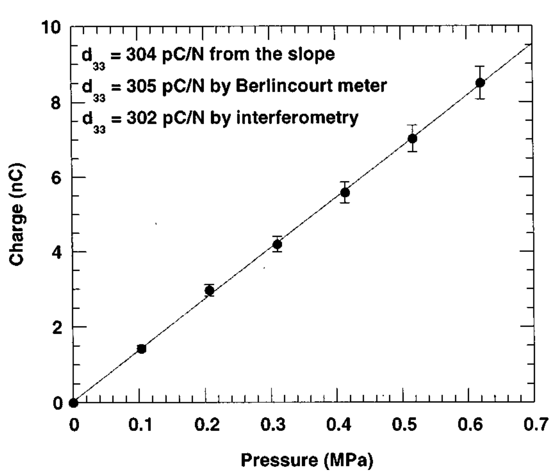


FIG. 6. Charge induced in a bulk PZT-5A ceramic specimen as a function of applied pressure. Measurements were made with the remnant in-plane stress self-compensation method.

B. Measurement on PZT thin films

The charge response of PZT thin films as the cavity pressure was changed was very similar to that observed in bulk samples (Fig. 2), indicating that the stress state applied to the thin film was the same as that for the bulk sample. Using the RIPSSC method described in the previous section, the d_{33} coefficient of several PZT thin films was measured. Figure 8 shows the induced charge as a function of the pressure change for a 1 μm PZT film poled at 150 kV/cm for 1 min. The relative dielectric constant and the remanent polarization of the film were 930 and 22 $\mu C/cm^2$, respectively. From the slope of the line, the effective d_{33} value of the thin film was

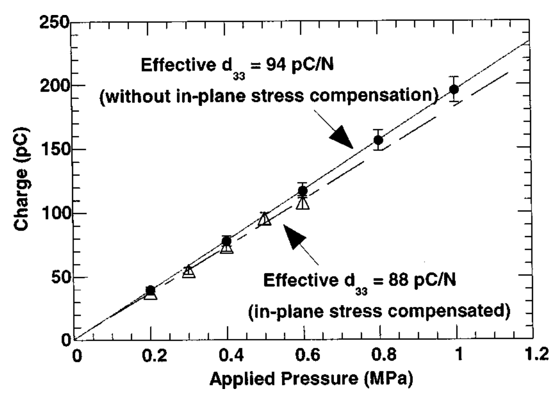


FIG. 8. Induced charge as a function of pressure change for a 1 μm PZT film poled at 150 kV/cm for 1 min.

TABLE I. Elastic properties of PZT film and silicon substrate (see Refs. 16 and 17).

S_{11}^E PZT (10^{-12} m^2/N)	S_{12}^E PZT (10^{-12} m^2/N)	S_{13}^E PZT (10^{-12} m^2/N)	$Y_{\text{Si}\langle 100\rangle}$ (GPa)	$Y_{\text{Si}\langle 110\rangle}$ (GPa)	Y_{ave} (GPa)	$\nu_{\text{Si}\langle 100\rangle}$	$\nu_{\text{Si}\langle 110\rangle}$	ν_{ave}
13.8	−4.07	−5.80	130	169.5	149.7	0.28	0.064	0.172

calculated to be 88 pC/N. Again a 7% error in the effective d_{33} value occurred when the RIPSSC method was not used.

Since double-beam interferometry is currently the most widely accepted technique for thin film d_{33} measurement and most of the d_{33} values reported in the literature were measured by that method, a direct comparison between the d_{33} value from our method and interferometry is necessary. The same electrode used for the data in Fig. 8 was measured using double beam interferometry. An effective d_{33} value of 84 pC/N was obtained. Again the two techniques gave very close d_{33} values, further confirming the validity of the charge measurement technique.

Given the fact that the RIPSSC method and interferometry yielded almost the same d_{33} value for a bulk PZT sample, it is our belief that the small difference in the d_{33} values obtained for the thin film was due to the different boundary conditions under which the two measurements were made. As was mentioned before, thin film piezoelectric measurements yield an effective value of the piezoelectric coefficient, due to the clamping effect of the substrate. The effective d_{33} value of a thin film on a substrate is related to the unclamped d_{33} value via the elastic and piezoelectric constants of the film and the substrate in a manner which depends on the boundary conditions. Since the boundary conditions for the direct piezoelectric measurement used here and a converse piezoelectric measurement such as interferometry are quite different, different effective d_{33} values are expected even when the same film is measured by the two techniques. Lefki and Dormans analyzed the boundary conditions for these two situations and predicted that the effective value given by the direct piezoelectric effect should be larger than the effective value given by the converse piezoelectric effect.[15] Our results agree with this prediction and are the first direct experimental observation of the influence of boundary conditions on the effective d_{33} of piezoelectric thin films.

To quantitatively evaluate the difference in the effective d_{33} measured by direct and converse methods, the model developed by Lefki and Dormans was adopted as a first approximation. In this model, the in-plane strain of the film was taken to be zero (assuming the film was totally clamped in-plane by the substrate) for the converse piezoelectric measurement, and it was equal to the in-plane strain of the substrate (assuming the substrate was free to expand and isotropic) for the direct piezoelectric measurement. The effective d_{33} for these two boundary conditions are given by[15]

$$d_{33}'(cp) = d_{33} - 2d_{31}s_{13}^E/(s_{11}^E + s_{12}^E), \tag{2}$$

$$d_{33}'(dp) = d_{33} - 2d_{31}(s_{13}^E + \nu/Y)/(s_{11}^E + s_{12}^E). \tag{3}$$

Here $d_{33}'(cp)$ and $d_{33}'(dp)$ represent the effective d_{33} measured by the converse and direct piezoelectric effect, respectively, s_{ij}^E and d_{ij} are the compliances and piezoelectric coefficients of the thin film, and Y and ν are Young's modulus and Poisson's ratio of the substrate. Because of the lack of compliance data for PZT thin films in the literature, data for an undoped bulk PZT ceramic with the same Zr/Ti ratio was used as an approximation.[16] For the calculation, the Young's modulus and Poisson's ratio of the substrate were taken as the average of the values in the ⟨100⟩ direction and ⟨110⟩ direction, which are the maximum and minimum in-plane values for a (100) silicon wafer.[17] Table I gives the values used for this work. Substituting them into Eqs. (2) and (3) yields:

$$d_{33}'(cp) = d_{33} + 1.19d_{31}, \tag{4}$$

$$d_{33}'(dp) = d_{33} + 0.96d_{31}. \tag{5}$$

Subtracting Eq. (4) from Eq. (5) yields

$$d_{33}'(dp) - d_{33}'(cp) = -0.23d_{31}. \tag{6}$$

Since the typical d_{31} value for these PZT films was around −40 to −50 pC/N,[18] the difference in effective d_{33} measured by direct piezoelectric effect and converse piezoelectric effect was estimated to be around 9–10 pC/N using Eq. (6).

However, in the real measurements, the boundary conditions deviate somewhat from the conditions used in the above model. Experimental results from single beam interferometry measurements indicate that the substrate is bent by the piezoelectric thin film during electrical excitation,[10,11] making the assumption that the in-plane strain was zero during the converse piezoelectric measurement invalid. In addition, during the direct piezoelectric measurement the substrate was not free to expand parallel to the plane because the periphery of the substrate was not subjected to the pneumatic pressure. Thus, the model represents two extremes for the boundary conditions and gives a maximum for the difference in effective d_{33}. The discrepancies between the measurements for the two techniques should and do fit between these bounds.

Kholkin *et al.* also used the above model to explain the difference between the d_{33} coefficients of bulk ceramic and thin film in PZT and Ca-modified lead titanate (PCT).[19] The d_{33} coefficient was significantly smaller in PZT thin films than that in PZT ceramics with the same composition, while in PCT, thin films and bulk material gave very similar d_{33} values. This was attributed to the effect of the boundary conditions on the d_{33} coefficient obtained in a converse piezoelectric measurement. For thin films, the effective d_{33} value is related to both the unclamped d_{33} and d_{31} of the piezoelectric material in a way similar to what is described in Eq. (4). However, PCT is a very anisotropic material and its d_{33} coefficient is about 20 times larger than the d_{31} coefficient.

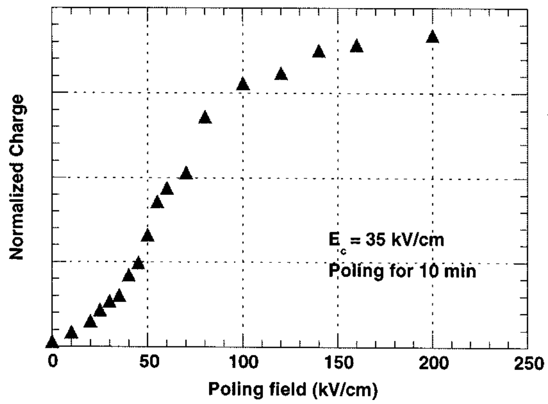


FIG. 9. Induced charge of a 1 μm PZT 52/48 film as a function of poling field.

Therefore, the difference in the unclamped d_{33} and the effective d_{33} in PCT is negligible, while it is significant in PZT due to the larger d_{31}.

In this work the d_{33} of the as-deposited PZT thin films was also measured and values between 0 and 10 pC/N were obtained. This result indicated that there was little or no preexisting alignment of the domains in the as-deposited films although all the films had strong preferred crystallographic orientation.[13] The dependence of the induced charge on the poling field was studied using another 1μm PZT film with (111) preferred orientation (Fig.9). The coercive field of this sample was about 35 kV/cm. As expected, a considerable increase of the induced charge was observed when the poling field exceeded the coercive field of the film. It started to saturate at a poling field of about three times the coercive field. Poling in the opposite direction led to a reversal in the sign of the induced charge, but the d_{33} value was almost the same for the two poling directions.

IV. CONCLUSIONS

(1) A measurement technique which could accurately measure the d_{33} value of bulk and thin film piezoelectric materials was developed. Good agreement among this technique, the Berlincourt method and double beam interferometry was obtained.

(2) Surface friction between the sample and O-ring was the major source of error. By optimizing the design and operation of the pressure system, error from the surface friction could be reduced to less than 5%.

(3) The measurement error can be eliminated using the RIPSSC method and an accurate d_{33} value could be obtained even if there was an appreciable amount of friction present.

(4) As-deposited sol-gel 52/48 PZT thin films showed very weak piezoelectricity with a d_{33} value less than 10 pC/N. The d_{33} value increased with poling field and saturated when the poling field exceeded three times the coercive field. Typical d_{33} values at saturation for 1 μm films were about 100 pC/N.

(5) A direct comparison between the d_{33} values measured by double-beam interferometry and this technique indicated the influence of the mechanical boundary conditions on the effective d_{33}. The effective d_{33} measured by the direct piezoelectric effect was slightly larger than that measured by the converse piezoelectric effect. A quantitative evaluation was made and the calculation was in agreement with the experimental result.

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