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Published on: 17 Mar 2004 - Journal of Physics D (IOP Publishing)

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Experimental investigation into the effect of substrate clamping on the piezoelectric behaviour of thick-film PZT elements

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Received 4 November 2003

Published

Online at stacks.iop.org/JPhysD/37

Abstract

This paper details an experimental investigation of the clamping effect associated with thick-film piezoelectric elements printed on a substrate. The clamping effect reduces the measured piezoelectric coefficient, d_{33} , of the film. This reduction is due to the influence of the d_{31} component in the film when a deformation of the structure occurs, by either the direct or indirect piezoelectric effect. Theoretical analysis shows a reduction in the measured d_{33} of 62%, i.e. a standard bulk lead zirconate titanate (PZT)-5H sample with a manufacturer specified d_{33} of 593 pC/N would fall to 227.8 pC/N. To confirm this effect, the d_{33} coefficients of five thin bulk PZT-5H samples of 220 μm thickness were measured before and after their attachment to a metallized 96% alumina substrate. The experimental results show a reduction in d_{33} of 74% from 529 pC/N to 139 pC/N. The theoretical analysis was then applied to existing University of Southampton thick-film devices. It is estimated that the measured d_{33} value of 131 pC/N of the thick-film devices is the equivalent of an unconstrained d_{33} of 345 pC/N.

1. Introduction

Bulk lead zirconate titanate (PZT) piezoceramics have many applications as both sensors and actuators [1] but are of limited use in micro-electro-mechanical systems (MEMS) because they are generally not compatible in terms of size or processing methods. Thin-film PZT layers are an alternative, but they are limited by the complexity of the deposition processes and the need to pattern the films afterwards. Screen printable piezoceramics, first reported by Baudry [2], allow relatively thick piezoceramic layers to be deposited directly onto the device. The thick-film process is additive, and hence the piezoceramic layers can be printed to a desired pattern without the need for subsequent etching. However, this process can introduce problems because the printed layer requires high temperature firing, 600–1000°C, to form a sintered piezoceramic layer. Other drawbacks include the limits of the resolution of the screen, typically 100 μm , and the ability of the device to withstand an applied printing pressure.

The boundary conditions applied to a piezoelectric element also influence piezoelectric behaviour. Optimum

piezoelectric behaviour is obtained when the piezoceramic is unconstrained in all directions. Thick-film piezoelectric elements, however, are rigidly clamped on one face to a substrate. This constraint limits the movement of the PZT layer as it attempts to expand or contract when a force or voltage is applied. Previous work [3, 4] has investigated the internal stresses and boundary conditions for thin-film ferroelectrics but has not related these effects to bulk devices and the reduction in piezoelectric properties observed. This paper presents, for the first time, an experimental comparison of both constrained and unconstrained bulk PZT-5H samples and relates the analysis to thick-film piezoelectric layers. The results are compared with theoretical analysis presented in [5–7] and applied to thick-film devices to determine the amount by which the clamping effect reduces the effective d_{33} coefficient of the printed PZT layer.

2. Theoretical evaluation of the clamping effect

The direct and indirect methods of measuring the piezoelectric coefficient are obtained from the relationships between charge,

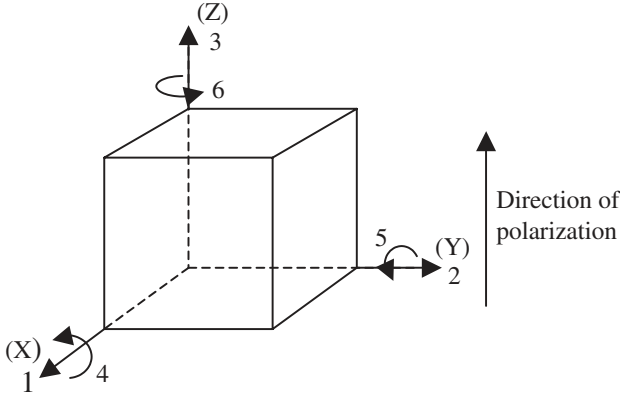


Figure 1. Notation of crystal axes with polarization direction.

induced strain and induced stress on the piezoceramic [8]. Equations (1) and (2) are defined as the piezoelectric constitutive equations:

$$S_{\alpha} = s_{\alpha\beta}^E T_{\beta} + d_{i\alpha} E_i \quad (1)$$

$$D_i = d_{i\alpha} T_{\alpha} + \varepsilon_{ij}^T E_j \quad (2)$$

$$\alpha, \beta = 1, 2, \dots, 6 \quad i, j = 1, 2, 3$$

where S , T , D and E are the strain, stress (N m^{-2}), electric displacement (C m^{-2}) and electric field (V m^{-1}), and s , d and ε are the compliance ($\text{m}^2 \text{N}^{-1}$), piezoelectric constant (C m^{-1}) and dielectric constant (F m^{-1}), respectively. The superscripts E and T indicate a constant electric field and constant stress, respectively. Figure 1 shows the orientation of the ceramic defined by α , β , i and j .

Equations (3), (4) and (5) are obtained from expanding the tensor notation.

$$S_1 = s_{11}^E T_1 + s_{12}^E T_2 + s_{13}^E T_3 + d_{31} E_3 \quad (3)$$

$$S_3 = s_{13}^E (T_1 + T_2) + s_{33}^E T_3 + d_{33} E_3 \quad (4)$$

$$D_3 = \varepsilon_3^T E_3 + d_{31} (T_1 + T_2) + d_{33} T_3 \quad (5)$$

2.1. Inverse effect

The inverse effect relates the piezoelectric coefficient, d_{33} , to the change in dimensions of a piezoceramic due to an applied field. The strain, S_3 , can be used to obtain the relation to d_{33} .

With no applied stress, $T_3 = 0$, the ratio of change in length (Δl) to the applied voltage (V) can be related to the strain and applied field.

$$\begin{aligned} \frac{\Delta l}{V} &= \frac{S_3}{E_3} = \frac{d_{33} E_3}{E_3} + \frac{s_{13}^E (T_1 + T_2)}{E_3} + \frac{s_{33} T_3}{E_3} \\ \rightarrow \frac{\Delta l}{V} &= d_{33} + \frac{s_{13}^E (T_1 + T_2)}{E_3} \end{aligned} \quad (6)$$

We are only concerned with piezoceramics that are mounted on a substrate; therefore it is necessary to consider the clamping effects on the piezoceramic as it attempts to change dimensions in axes 1 and 2. Assuming that the substrate has greater rigidity and relative thickness than the piezoceramic allows the assumption that the displacement in

axes 1 and 2 is negligible and therefore the strain $S_1 = S_2 = 0$. We also assume that the substrate is the dominant element and that it is isotropic and therefore $T_1 = T_2 = T$. Using equations (3) and (4), it is possible to express d_{33} independent of T .

$$\begin{aligned} S_1 &= s_{11}^E T_1 + s_{12}^E T_2 + s_{13}^E T_3 + d_{31} E_3 \\ \therefore -d_{31} E_3 &= (s_{11}^E + s_{12}^E) T \rightarrow \frac{T}{E_3} = \frac{-d_{31}}{(s_{11}^E + s_{12}^E)} \quad (7) \\ \therefore \left(\frac{\Delta l}{V} \right)_T &= d_{33(\text{eff})} = d_{33} - 2d_{31} \frac{s_{13}^E}{(s_{11}^E + s_{12}^E)} \end{aligned}$$

Thus equation (7) gives the effective d_{33} value that is measured. From this, the unconstrained d_{33} value can be determined.

2.2. Direct effect

The direct effect concerns the amount of charge generated when a force is applied to the piezoceramic. If a bar is subjected to a stress, it will expand and contract along its axes depending on the direction of the stress. The lateral strains in the device are related to the stress applied along axis 3, denoted as T_3 . Assuming an isotropic substrate that dominates the piezoceramic, the lateral strain is the same in each direction and therefore $S_1 = S_2$ and the stress $T_1 = T_2 = T$ due to the substrate clamping. This is different from the indirect effect because the applied stress is transferred via the film to the substrate. Thus, the values of S_1 and S_2 must be determined as a function of T_3 using Hooke's law, shown in equation (8), where ν_{sub} and Y_{sub} are Poisson's ratio and Young's modulus of the substrate, respectively.

$$S_1 = S_2 = - \left(\frac{\nu_{\text{sub}}}{Y_{\text{sub}}} \right) T_3 \quad (8)$$

Relating the strain and the ratio of the charge generated to the applied force and combining this with equations (3), (4) and (8) gives equation (9).

$$- \left(\frac{\nu_{\text{sub}}}{Y_{\text{sub}}} \right) T_3 = (s_{11}^E + s_{12}^E) T + s_{13}^E T_3 + d_{31} E_3 \quad (9)$$

For the short circuit condition, $E_3 = 0$, the assumptions made concerning the direct effect can be combined with equations (5) and (9) to obtain equation (10).

$$\left(\frac{D_3}{T_3} \right)_E = \left(\frac{2d_{31} T + d_{33} T_3}{T_3} \right)_E \quad (10)$$

Re-arranging equation (9) and substituting into equation (10) gives the effective d_{33} , denoted $d_{33(\text{eff})}$ for the direct effect (as shown in equation (11)).

$$\left(\frac{D_3}{T_3} \right)_E = d_{33(\text{eff})} = d_{33} + 2d_{31} \left(\frac{-(\nu_{\text{sub}}/Y_{\text{sub}}) - s_{13}^E}{s_{11}^E + s_{12}^E} \right) \quad (11)$$

The values for **PZT-5H type** materials were obtained from Morgan Electro Ceramics [1] and Efunda [9]. We have assumed a clamped film on a 96% pure alumina substrate, and therefore the Poisson's ratio and Young's modulus values for



the substrate will be used; these values were obtained from Hybrid Laser Tech [10].

$$\begin{aligned} \nu_{\text{sub}} &= 0.25 & Y_{\text{sub}} &= 331 \text{ GPa} \\ s_{11}^E &= 16.4 \times 10^{-12} \text{ m}^2 \text{ N}^{-1} \\ s_{12}^E &= -4.78 \times 10^{-12} \text{ m}^2 \text{ N}^{-1} \\ s_{13}^E &= -8.45 \times 10^{-12} \text{ m}^2 \text{ N}^{-1} \end{aligned}$$

Substituting these values into equation (11) gives equation (12):

$$d_{33(\text{eff})} = d_{33} + 1.33d_{31} \quad (12)$$

From the Morgan Electro Ceramics data book, typical values for d_{33} and d_{31} are 593pC/N and -274pC/N , respectively. Therefore, using equation (12), the measured d_{33} values for clamped thick-film 5H-type PZT devices have a theoretical maximum of 227.8pC/N, a reduction of 61% from the bulk value. This formula shows that the actual d_{33} of the material remains the same but the effective measurement of that d_{33} is reduced.

3. Practical measurement of the clamping effect

Thick-film samples cannot be used to demonstrate equation (11) because it is impractical to remove the devices from the substrate for direct measurement of d_{33} . Therefore, thin bulk samples from the Morgan Electro Ceramics product range were attached to a substrate to observe the effects on d_{33} .

Five metallized bulk 5H-type PZT discs, average thickness 220 μm and diameter 9.6 mm, were obtained from Morgan Electro Ceramics and their d_{33} coefficient measured. These were fully processed bulk piezoceramics that had been previously fired and poled by Morgan Electro Ceramics. The five discs were de-poled by heating them above the Curie temperature on a belt furnace and then re-poled at the Morgan Electro Ceramics factory using a proprietary manufacturing process. The piezoelectric coefficient, d_{33} , was then measured for each disc as a reference point for the unclamped disc. The bulk samples were then bonded on 0.64 mm thick 96% alumina substrate. An ESL 9633B silver-palladium electrode was printed on the substrates and the bulk discs placed on the wet printed pastes. The samples were dried in a DEK 1209 IR drier and fired with an 890°C peak temperature furnace profile. The plane view of the assembled samples is shown in figure 2. The silver-palladium film provides an electrical connection to the bulk piezoceramic whilst simultaneously rigidly bonding it to the substrate. Because the furnace temperature is above the Curie temperature of 5H-type PZT, it was necessary to re-pole the discs once fired. The samples were re-poled at the Morgan Electro Ceramics factory with parameters identical to those used before firing to ensure consistency with the poling level and decay time, thereby enabling a direct comparison between the unclamped and clamped d_{33} . In addition, it ensured that any reduction in d_{33} would be known to be due to the clamping effects rather than the device not obtaining polarization saturation, which would also result in a lower d_{33} . The d_{33} values before and after clamping were measured using a Take Control PM35 piezometer; five measurements were taken for each sample with a frequency setting of 97 Hz.

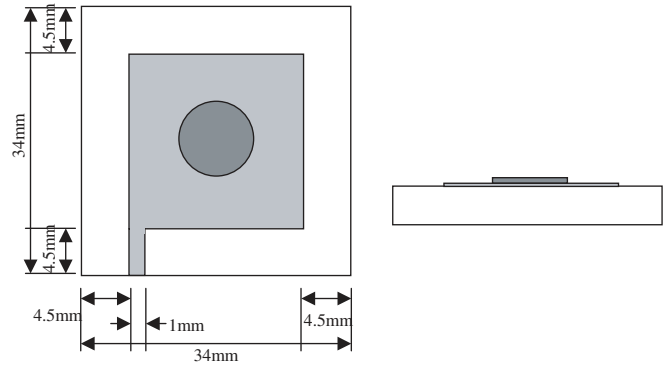


Figure 2. Sample design layout for substrate, electrode and PZT disc.

4. Experimental results

The values used for d_{31} in equation (11) were calculated as a ratio of the data sheet value for d_{31} compared with the measured value of d_{33} . This assumes that the d_{33} and d_{31} values change proportionally. These values were used to calculate the theoretical value of $d_{33(\text{eff})}$ using equation (12).

Figure 3 shows a comparison between the unclamped bulk devices, theoretical values given by equation (12) and the measured values once the disc was attached to the substrate and re-poled. These results show an average unclamped d_{33} of 529pC/N and an average clamped value of 138pC/N, giving a reduction of 74%.

5. Discussion of results

Figure 3 indicates that clamping a piezoceramic to a substrate reduces the measured d_{33} . The clamped 5H samples have $\sim 32\%$ lower measured d_{33} values than the theoretical values obtained from equation (12). This further reduction in d_{33} could be attributed to the lead loss experienced by the PZT layer when fired in the belt furnace. However, the firing temperature was only slightly above that at which lead migration occurs and therefore unlikely to have a large influence on the stoichiometry of the bulk device. This would indicate that any change in the domain structure of the bulk device would be minimal but could be a factor in the extra reduction observed.

Another contributing factor is that, despite the poling parameters before and after clamping being identical, the effect of the applied poling field after clamping is reduced. The electric dipoles within the ceramic cannot align to the same degree as an unclamped bulk ceramic when the poling field is applied because the associated deformation of the device is restricted by the substrate. This effect means that the polarization saturation point in the bulk device is reduced, thus causing an overall reduction in the piezoelectric activity. Another possible source for error is that the theoretical calculation ignores the shear effects within the film structure, which in practice could be introducing a further negative effect on the measured piezoelectric activity. Reductions in d_{33} due to grain size or size related domain structure transitions [11, 12] can be ignored because the bulk disc was already fully sintered before being clamped to the substrate, as evidenced by the high d_{33} measurements before clamping.

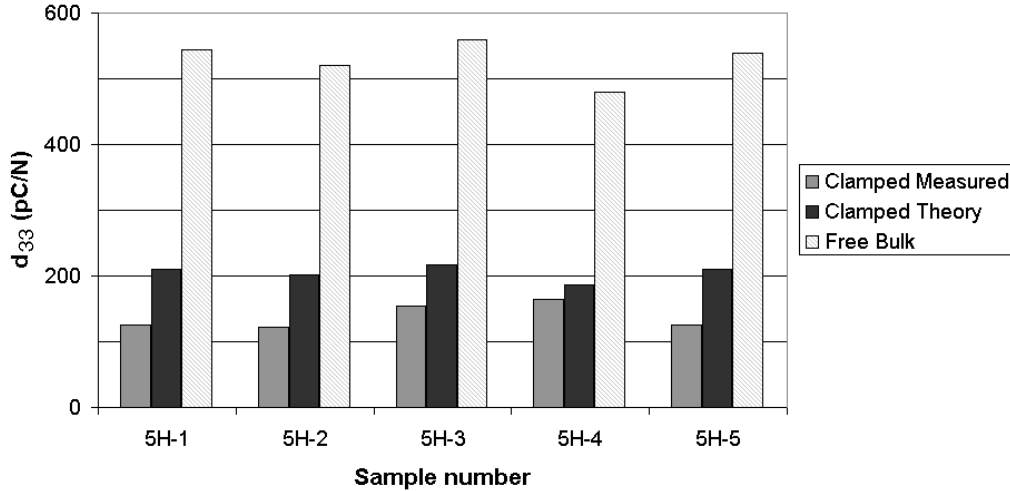


Figure 3. Comparison of theoretical and measured values of clamped d_{33} .

5.1. Thick-film samples

Using these results, it is possible to estimate the unconstrained d_{33} value from the $d_{33(\text{eff})}$ value measured for the University of Southampton thick-film devices, 131 pC/N, using equation (12). These devices represent a continuation of the optimization work concerning 5H-type PZT thick-film devices conducted at the University of Southampton [13]; they consist of a blend of ball and attritor milled PZT powders with a lead borosilicate binder fired at a peak temperature of 1000°C and with optimized poling parameters. The device comprises a square of PZT ($1.1 \times 1.1 \text{ cm}^2$) of 60 μm thickness printed on a standard 96% alumina substrate. It is not possible to measure the unclamped d_{31} for these devices, and therefore this is assumed to be the ratio of bulk d_{33} and d_{31} . A d_{31} value of -274 pC/N can be used in equation (12) to produce a theoretical estimate of 345 pC/N.

$$\frac{d_{33}}{d_{31}} = \frac{593}{-274} \rightarrow d_{31} = -\frac{d_{33}}{2.16}$$

$$d_{33(\text{eff})} = d_{33} - 1.33 \left(\frac{d_{33}}{2.16} \right) \quad (13)$$

$$d_{33} = 2.63 d_{33(\text{eff})}$$

$$d_{33} = 2.63(131 \text{ pC/N}) = 345 \text{ pC/N}$$

This is still less than the equivalent bulk 5H-type device with a d_{33} of 593 pC/N; this discrepancy will be due to the different domain and structural effects that the thick-film device is subject to as a result of its manufacture [11, 12].

6. Conclusions

This paper has confirmed that the addition of a substrate to a piezoceramic has significant effects on the measured d_{33} value. The clamping effect equation [5], given in equation (12), has been shown to be valid with the measured results showing a significant reduction in d_{33} as predicted. For the bulk 5H devices, a reduction of average values from 529 pC/N to 139 pC/N was observed. This was found to be an approximately 74% reduction in the measured d_{33} value.

This result is significant because it provides an indication of the maximum achievable d_{33} for a particular PZT type and substrate combination. This is important when considering which powder and process combinations to use when designing devices for a particular application. This is of direct relevance to work on thick-film devices, where the measured d_{33} value is considerably lower than that of bulk devices. These results give an estimation of the proportion of the reduction due to the clamping effect and that due to processing and material differences between bulk and thick-film devices.

Further work will investigate the effect of the top electrode on the overall clamping of the piezoceramic and how this effect can be reduced with the use of alternative electrode materials.

Acknowledgments

The authors would like to thank Morgan Electro Ceramics Ltd for their continued support and assistance.

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Summary of Comments on d171423

Page: 2

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PZT-5H type

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