Measurement Good Practice Guide No. 44

Measuring Piezoelectric d_{33} coefficients using the Direct Method

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Abstract:

Although piezoelectric materials are often used as actuators in order to make small precise movements it can be difficult to measure these displacements in an industrial environment. Consequently simpler methods have been sought to measure the piezoelectric activity, such as resonance methods, and measurement of the piezoelectric coefficient d_{33} using the direct method (often called the Berlincourt method).

This good practice guide will examine the advantages and disadvantages of the method in detail and with some experimental validation using typical PZT ceramics examine the validity of using the data from this method to predict the displacement of materials in real conditions.

ISSN 1368-6550

June 2001

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This guide was produced as part of the *Materials Measurement* programme, a programme of underpinning research supported by the Department of Trade and Industry and disseminated by the National Physical Laboratory.

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Contents

1. Simple Checks for Improved Measurements for Quasi Static Piezoelectric Coefficient (Berlincourt) Measurements

Static Pre-load : Think about the load applied to the sample. A 10N load on a 1mm area is 10MPa. Will this affect your sample? Check the effect by changing the pre load.

AC load level : Similarly with the AC load level, is this affecting your measurements? If you can change the level, do so, and look at the effect. Generally the lower AC level will give more linear behaviour, but at the expense of decreased signal to noise ratio.

Sample Geometry : Thicker samples are always better in terms of spreading the loads, a thinner sample might give increased stress levels. Check the effect on thinner samples by stacking several and looking at the difference. There should be no difference.

Measurement Frequency: Don't work exactly on mains frequency, or multiples thereof. Check the resonance behaviour of your system using either Lithium Niobate, or a very hard PZT composition. These should show little change with frequency. Work as far away from the resonance as possible. Work at the calibration frequency, if you want to change frequency, consider recalibration at this frequency.

Calibration : How was your reference sample calibrated, and is this calibration valid at the frequency and stress levels used in your system? Use a high sensitivity calibration sample to set the gain of the system, but check the gain independently with a lower activity reference material.

Environment : Understand the temperature compensation mechanism of your system, i.e. what is the temperature sensitivity of the internal reference? If you measure a sample with similar properties to the internal reference there will be no variation with temperature. Is this what you want? If the internal reference is a high sensitivity PZT material, check the temperature variation with a more temperature stable material.

What are the effects of moisture and air currents on your system? Look at the measurements as you breathe moist air onto it. Is it stable if you blow dry air over it, for instance from a compressed gas source?

Loading probes : Flatter loading contact geometries are better for thinner samples because they reduce the effective stress levels, and also ease misalignment problems. However using completely flat loading contact can lead to bending stresses unless all surfaces are perfectly aligned.

Time Dependant Effects : The time dependence of measurements is usually small but measurable. Leave a sample in the system overnight and see the difference. For consistency of results it is good practice to take readings after a fixed time, for example after 5 seconds.

2. Introduction

The piezoelectric charge coefficient, d_{ij}, is one of the fundamental parameters defining the piezoelectric activity of a material, basically the higher the d_{ij} the more active the material is. Consequently manufacturers, designers, and users want to know the d_{ij} coefficient for the material.

Measurement of the d_{ij} coefficient can be realised in several ways varying in accuracy and simplicity. The most reliable method of determining the d_{ij} coefficient is to electrically excite a resonance in a sample, and from the resonance given the dimensions of the sample, and the density a d_{ij} coefficient can be calculated. One problem with this method is that the geometry of the sample must be such that only a pure fundamental resonance mode is produced, and the calculated d_{ij} parameter relates to this resonance mode. This leaves the problem how to determine the d_{ij} parameter for shapes that don't have an ideal resonance geometry, or where the resonance mode is not the mode that will be used. For instance, for thin discs poled in the thickness direction it is easy to excite a resonance in the radial direction, and determine the relevant d_{ij} parameter, but to get the d_{ij} coefficient for motion in the thickness direction longer cylinders are needed.

The d_{ij} coefficient is defined as the charge produced for an applied stress, or the strain for an applied voltage, and these are theoretically equivalent. The latter measurement is more difficult to achieve because of the small strains involved, so measurement techniques have concentrated on the former. Initially the charge was measured in response to an applied static load, but difficulties with thermal drifts led to the measurements being performed quasi statically, at a few hundred hertz.

The quasi static method is simple; a small oscillating force is applied to the sample and the charge output is measured and divided by the applied force amplitude. The simplicity of the technique has been its downfall, in that anyone can easily build up their own system, and there are a growing number of commercial systems. There are currently no standards for this measurement method, and consequently each system performs the measurement slightly differently. This means that, although the results from these systems are good for measuring within a batch or batch to batch variability, external comparisons usually produce a large variability. This has led to a loss in confidence in these measurement results.

Interlaboratory tests have shown systematic differences between results from different laboratories. Figure 1 shows the results from a recent round robin with four participants, illustrating the behaviour for a soft PC 5H material. For comparison ±5% error bars have been drawn on the results for laboratory 2, and it can be seen that many of the results lie within this error band. There is evidence that the errors are systematic, i.e. partner 1 is almost always the lowest and partner 4 the highest. This suggests the measurement method could be improved by closely examining reasons for these deviations.

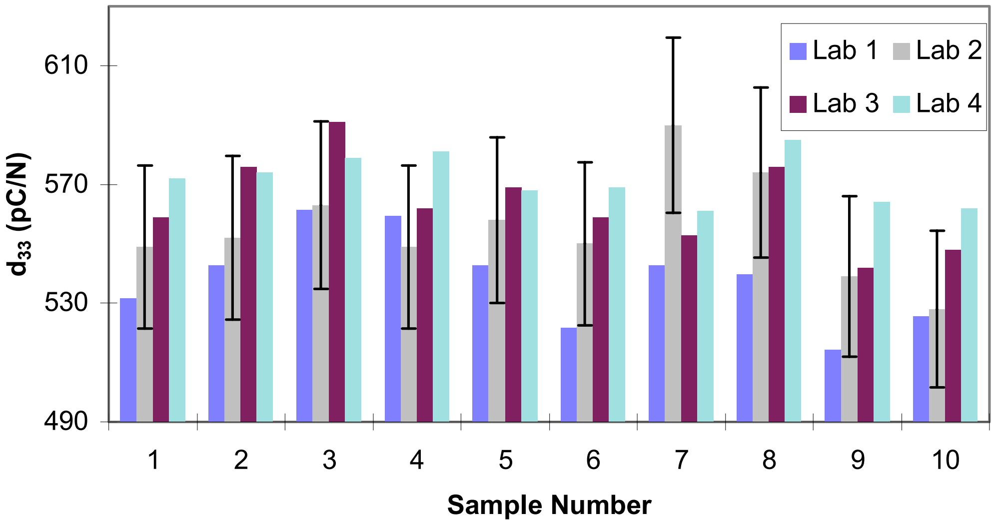

Figure 1 Round robin results on PC 5H material

This good practice guide aims to answer two questions. Firstly, is the Berlincourt test a valid test method? For example what are the parameters controlling the accuracy and reliability of the results? Secondly, does the Berlincourt test result predict what displacements can be achieved when the material is excited electrically?

2.1 History and Physical Basis for Quasi-Static d_{33} Measurement

The phenomenon of piezoelectric (from the Greek word "piezo" or "to press"), was discovered in 1880 during a systematic study of the effect of pressure on the generation of electric charge by crystals. Materials such as quartz and Rochelle salt were some of the crystal structures studied, and due to their non-centrosymmetric crystal structures (i.e. structures lacking a centre of symmetry to their unit cell) an applied stress resulted in the generation of electric dipoles, a process known as polarisation. The applied stress causes the ions in the unit cell to move from their equilibrium positions and distort relative to one another. The necessity for a non-centrosymmetric crystal structure can be rationalised when one considers that a homogenous externally applied stress is centrosymmetric and thus cannot produce a non-centrosymmetric result, such as a vector-quantity like polarisation, unless the material lacks a centre of symmetry.

Such piezoelectric materials also exhibit the converse piezoelectric effect whereby a mechanical deformation is induced on application of an external electric field. The generation of a surface charge on deformation is known as the direct or generator effect, whereas the converse effect of deformation on application of an electric field is designated the motor effect.

The performance of piezoelectric ceramic materials can be quantified either statically (under the influence of a steady strain), quasi-statically or dynamically using the resonance method. The earliest piezoelectric materials investigators determined the piezoelectric constants using static tests, but, due to the difficulty in controlling electrical boundary conditions, static test are now seldom used for piezoelectric materials. Dynamic performance relates to the behaviour of a material when subject to an alternating electric field or mechanical stress applied at frequencies close to the mechanical resonance of the component. The dynamic method, and to a lesser extent the quasi-static method, eliminates the drift due to pyroelectric charges that produce errors in static measurements, and is thus considered to be of higher precision. Additionally, the utilisation of an alternating field allows a more convenient measurement than that carried out at DC. It must be noted that the frequency of the applied signal must be less than the fundamental resonant frequency of the sample and its mounting in order for the equations applied to the static measurements to remain valid and for accuracy to improve in this way.

A means of quantifying piezoelectric performance is by use of the piezoelectric equations and coefficients. These describe the interaction between electrical and mechanical behaviour and can be summarised as follows:

"*d*" and "*g*" are the charge and voltage piezoelectric constants respectively, related by the expression:

$$d = \varepsilon_r \varepsilon_0 g$$

where,

ε_r = relative permittivity (dielectric constant)
ε_0 = permittivity of free space (8.85 $pF.m^{-1}$)

Interestingly, the difference between *d* and *g* is electrically related to short and open circuit conditions for the piezoelectric materials, respectively.

In summary, the piezoelectric coefficients are defined as follows:

Direct Effect	Converse Effect
$d = \frac{\text{charge density developed}}{\text{applied mechanical stress}}$	$d = \frac{\text{strain developed}}{\text{applied field}}$
$g = \frac{\text{electric field developed}}{\text{applied mechanical stress}}$	$g = \frac{\text{strain developed}}{\text{applied charge density}}$

Due to the anisotropic nature of poled ferroelectric ceramics (materials with artificially induced and enhanced piezoelectric properties), and the freedom with which one can select the poling direction, it is necessary to formalise a method for identifying the axes of a component in order to specify its parameters.

The direction the material is poled is conventionally taken as the 3-axis (z) with the 1 and 2 axes being perpendicular. Thus, it follows that the subscripts 4,5 and 6 refer to shear strains associated with the 1, 2 and 3 directions, Figure 2.

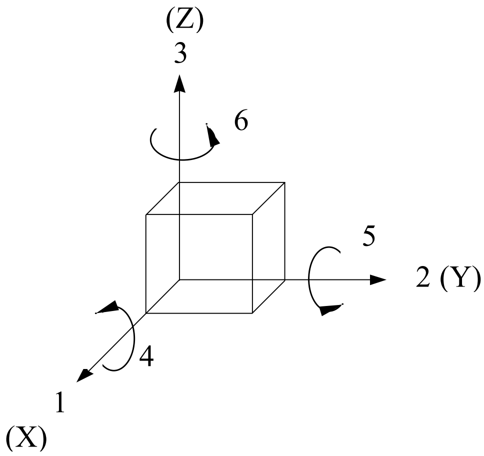

Figure 2: Conventional Axis Notation For Ferroelectric Crystals

2.2 Mathematical Basis for Measurement Method - Piezoelectric Relations

The Piezoelectric effect is an interaction between the mechanical and electrical behaviour of the material, and to a linear approximation, this interaction can be described by the expressions:

$$S = s^{E}T + dE \qquad (1)$$

$$D = \varepsilon^{T}E + dT \qquad (2)$$

Where

E	=	Electric Field Strength
D	=	Dielectric Displacement
T	=	Applied Stress
S	=	Strain
s	=	Compliance
ε	=	Permittivity

The superscripts E and T are used to indicate values determined at constant field and stress respectively.

The above equations are a generalised form, without the directional notation. If we consider the common case of a ceramic poled in the 3 direction, with electrodes on these faces, and we maintain a constant field, i.e. at zero, then equation 2 becomes:

$$d_{33} = [\delta D_3 / \delta T_3]_E \qquad (3)$$

where, D = electric displacement in the 3-direction (z)
T = applied stress also in the 3-direction (z)

With reference to the measurement of d_{33}, equation 3 can be rewritten as:

$$d_{33} = [(Q/A).(F/A)] = (Q/F) \tag{4}$$

Where, F = Applied Force
A = Area force applied over
Q = Charge developed

The importance of equation 4 is that it gives us a method of determining d_{33} using the direct method, by applying a force on a piezoelectric sample and measuring the charge developed. The areas cancel out so there is no need to measure the area. The only proviso is that the measurement must be performed at a constant field, i.e. under short circuit conditions. This can be achieved by measuring the charge using a virtual earth amplifier or by ensuring that the Device Under Test (DUT) is effectively short circuited by placing a large enough capacitor (much greater than the capacitance of the DUT) across the DUT.

Equation 1 can also be rewritten as

$$d_{33} = [\delta S_3 / \delta E_3]_T \tag{5}$$

so that now the experiment is performed at a constant stress. This gives us a means of measuring the d_{33} in the indirect mode, that is application of a field to produce a strain, all at a constant stress.

This gives units in the direct mode of C/N and the indirect m/V, and for PZT materials is usually in pico units. The units are equivalent.

2.3 Measurement Apparatus - Theory of Operation

A piezoelectric coefficient meter consists of two parts, the force head, and the control electronics.

The force head incorporates the loading actuator, reference sample, and some means of accommodating different sized samples. The method of applying the oscillating force is usually via a loudspeaker type coil, as this is a relatively cheap and simple means of delivering the force to the sample. The reference sample is in line with the loading train and is used to measure the force applied, so that the reference sample experiences the same loads as the DUT. The reference sample is often a PZT ceramic in order to provide high sensitivity and also a crude form of temperature compensation. The clamping of the samples in the system is usually achieved through a screw thread adjusted vice that forces the sample against the loading actuator. The coil actuator is held in place by some sort of leaf spring arrangement and the stiffness of this spring and the number of turns on the clamping system determines the pre load put on the sample. This pre load is required to achieve stable measurements so that the sample does not rattle. Figure 3 shows a schematic of the components in the force head.

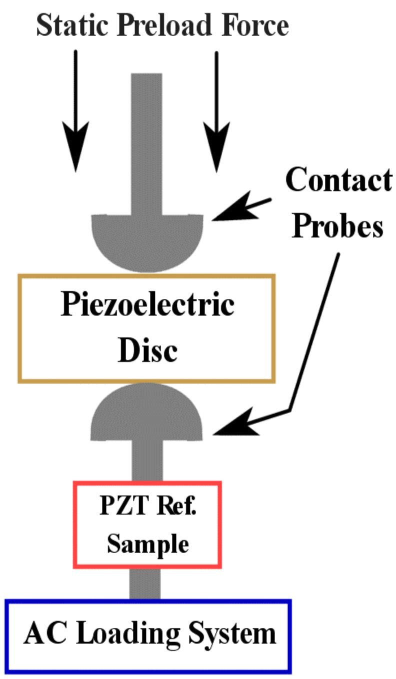

Figure 3: Schematic of Force Head

The control electronics provide the amplified AC signal to drive the coil at the required frequency, the charge measurement system, the d_{33} calculation system and the readout electronics. The frequency range of these systems are limited at the lower frequencies to around 10Hz by thermal drifts, and at the higher end, around 1kHz, by the mechanical resonance of the complete force head assembly. Sometimes the frequency and the applied force can be selected, although the measurements should be insensitive to the applied AC load for the levels used in this experiment (but see later).

The charge measurement electronics must be such that the experiment is carried out under constant field conditions, so either the current is measured using a virtual earth amplifier or the current is shunted through a capacitor of capacitance orders of magnitude greater than the sample (short circuit conditions). The d_{33} measurement is the ratio of charge from the reference material (the force) to the charge developed in the DUT. Either the Root Mean Squared (RMS) values of these signals can be measured and divided to give d_{33} or the two signals could be divided electronically by using a differential amplifier and then the RMS value of this difference signal will be proportional to the d_{33}.

The RMS signals are proportional to the d_{33} and the electronic control system must provide some means of calibration to return the correct value. Usually this is achieved by placing another reference calibration sample as the DUT and then the displayed value is adjusted to give the certified value of the calibration sample.

2.3.1 "The Berlincourt Method"

The use of the name "The Berlincourt Method" is somewhat of a misnomer when referring to quasi-static measurement of the piezoelectric d_{33} coefficient using the direct effect, as the skeletal basis for the measurement schema was proposed in "Piezoelectric Ceramics" by B. Jaffe, W. R. Cook Jr. And H. Jaffe. The commonly used name refers to the scientist, Don Berlincourt, who was working for Channel Products during the development of one of the first commercial d_{33} measurement systems. Similar systems are also termed piezometers, d_{33} testers, or piezo d meter.

3. Experimental Investigation of a Berlincourt System

3.1 AC measuring force

The level of the applied AC measuring force can be varied for some systems. The magnitude of the force is not usually important, as long as the stresses introduced into the material mean the piezoelectric is still operating in the linear regime. Typical operating AC loads are round 0.1N RMS. If the force can be varied then this can be investigated to check that increasing the AC force has no effect on the measured d_{33}. The obvious advantage of using increased loads is that a larger charge signal will be generated, and this can reduce the signal noise related errors, however the lower the applied force the greater the likelihood that the material will behave linearly. The level of the AC force can be checked by inserting a load cell in the measuring head, a quartz load cell is useful for providing adequate dynamic response.

3.2 Effect of Frequency

The frequency range of the Berlincourt type instrument is limited to a range between roughly 10Hz to 1kHz. The lower frequency limit is governed by the charge measurement system, and the stability of the charge over the measurement period. Problems with thermal drift and charge dissipation through the surroundings are one reason that the measurements are not performed at DC (static). The upper frequency limit is governed by the load application method which is based upon a loudspeaker drive coil, and the ability of the system to deliver useful stresses into the sample. Also, it is difficult to design a load application that does not display a resonance peak that interferes with the measurement range. The system is designed so that the peak is far enough away that the measurements are made in a frequency independent region.

Because the method is quick and simple it is not usual to completely shield the instrument electrically from the surroundings, and so there are often measurement anomalies at the mains power frequencies and its corresponding harmonic frequencies. Consequently, it is usual to work just off one of these frequencies, say at 97Hz.

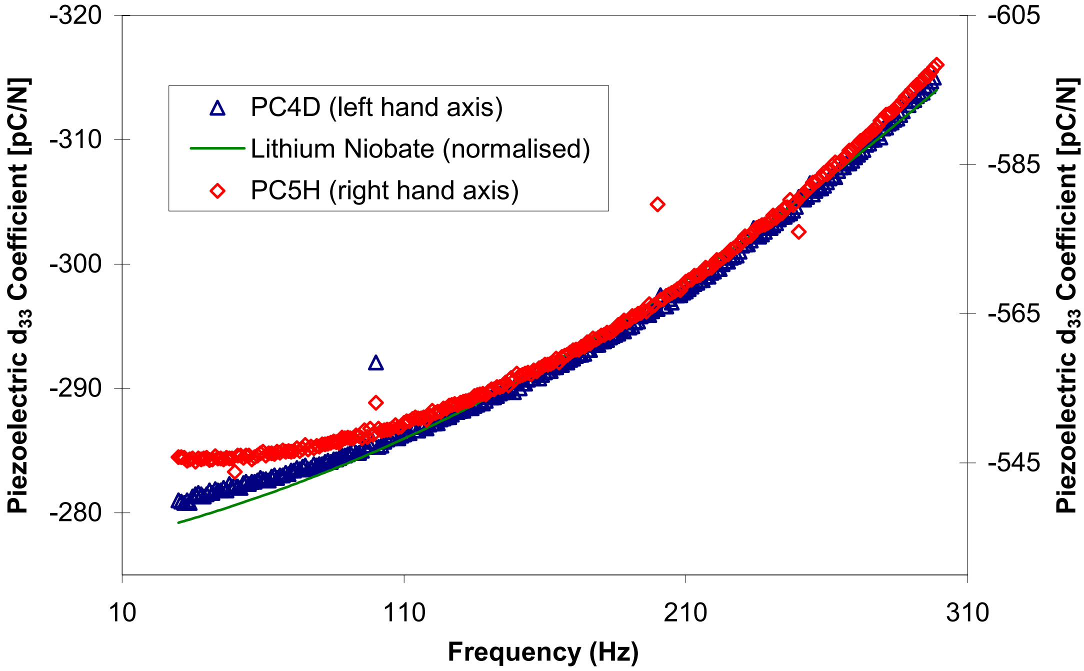

Figure 4: Frequency sweep for PC4D, PC5H and lithium niobate

Figure 4 illustrates some of the issues related to the frequency response of the system, and the effect in different materials. The trace for the PC 4D material shows measurement outliers at 50, 100 and 200Hz as discussed in the preceding paragraph. To illustrate the material's dependency, the results for the hard and the soft material have been plotted on separate Y axes, and a third line for a fit of measured values on Lithium Niobate has been normalised in a similar manner. Lithium Niobate has a d_{33} of around 20, and is thought to behave linearly with respect to applied fields etc. Measurements using the Berlincourt meter on a $LiNbO_3$

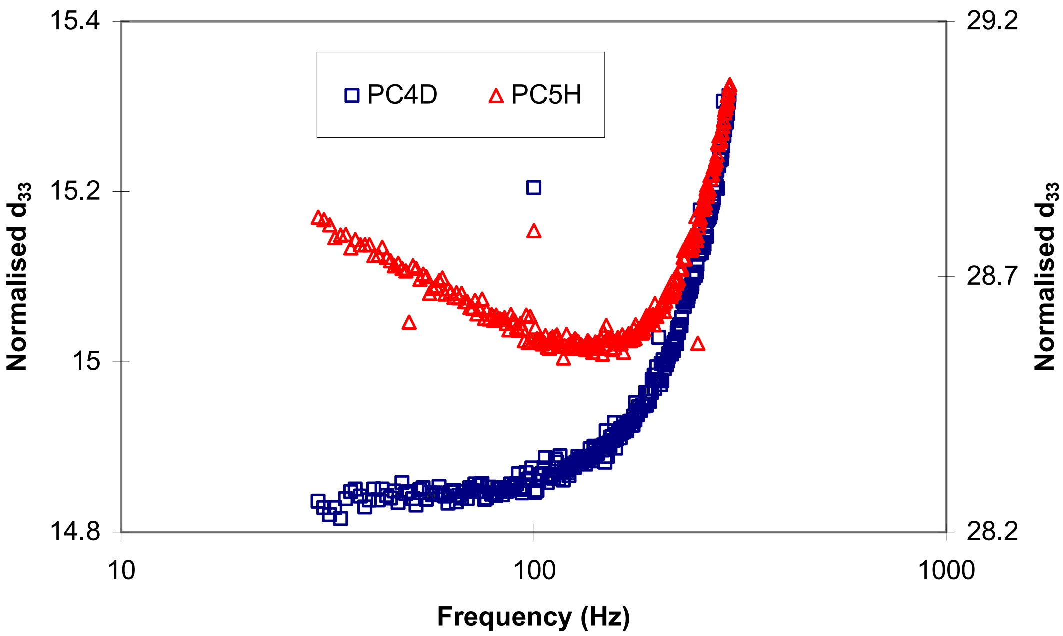

Figure 5: Frequency Sweep for PC4D and PC5H normalised using lithium niobate

single crystal increased from 19 to 21.5pC/N over the frequency range 30 - 300Hz. We can therefore tentatively assume that this is due to the system response and is increasing towards a resonance peak in the kHz region. In contrast, the soft material displays a pronounced up-turn in the results at low frequencies, whilst mirroring the behaviour of the PC 4D and $LiNbO_3$ at frequencies above 150Hz. This up-turn must therefore be material dependent, i.e. that the d_{33} increases with decreasing frequencies for the soft material. This effect has been seen in various experiments on soft piezoelectric materials and is due to the inhibition of domain movement at increasing frequencies. This behaviour has been modelled by the Rayleigh law, due to the parallels in between ferromagnetic and ferroelectric materials. One of the observations for materials systems controlled by domain wall motion is a logarithmic dependence of behaviour with respect to frequency. The Rayleigh model is often applied to soft ferroelectric materials when they are operating in a region where domain wall movement is dominant, i.e. at large stresses and for low frequencies. If the results for the hard and soft material are divided by those for the lithium niobate at each frequency then this should effectively cancel out any system dependant effects. This has been done in Figure 5 and plotted against the log of the frequency. Both samples show a similar increase above around 100Hz, but below this the hard material is largely frequency independent, whereas the soft material shows a linear increase with decreasing frequency. The behaviour below 100Hz is consistent with known behaviour, however above this it would appear that it is still system dependant, since the behaviour for hard and soft is identical. It may be that because the system gain is set by calibration at 100Hz then the gain is valid here but at the higher frequencies there may be a frequency dependant gain issue.

3.3 Effect of Static Pre load

Samples are clamped in position by a small pre-load which is usually applied by turning a screw thread on the measuring head. This forces the sample against some sort of spring, and the stiffness of this spring and the number of turns on the screw thread governs the value of the pre load. The reason for the pre load is simply to hold the sample in position while the

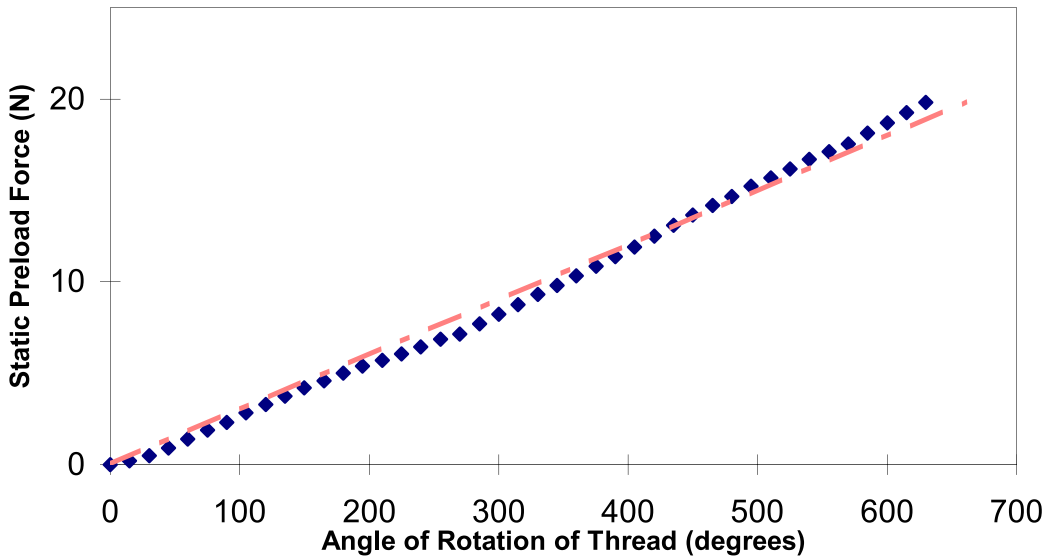

Figure 6: Calibration of static pre-load. One turn exerts 10.8 N force.

oscillating measuring force is applied. The pre load need only be greater than half the amplitude of the measuring load, i.e. so that the probes never leave the sample, but in practice the value is much greater than this.

Figure 6 shows the linearity of the pre load with number of turns of the preload mechanism as measured by a small strain gauged load cell. The slope of this curve is effectively the stiffness of the pre load spring. From this it can be seen that the value of the applied pre load might be expected to vary around ± 10% in typical tests.

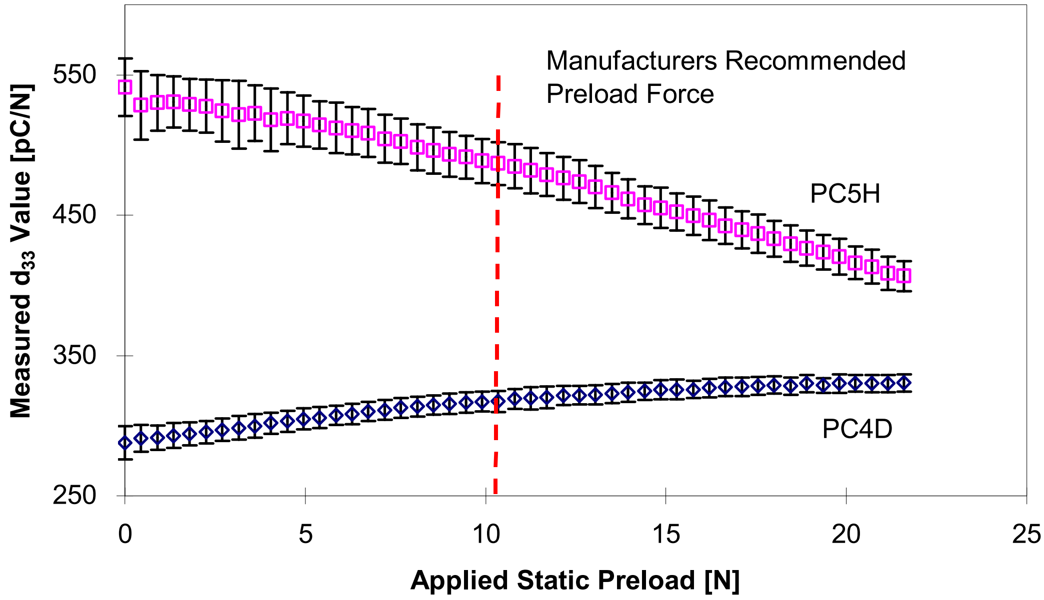

Figure 7: Effect of static pre load on PC 4D and PC 5H

The value of the pre load to use in these type of instruments was set historically, based on a value that gave the most consistent results, which is a compromise between enough force so that the sample does not rattle, and not enough to completely *clamp* the material. That the pre load can have an effect on the measurements can be seen in Figure 7. Here the static pre load is increased from 0 to over 20N on a typical hard and soft PZT material. For the soft material, the increasing pre load has the effect of reducing the d_{33} whereas for the hard material the behaviour is reversed. The slope of the soft material is also much greater showing that the pre load has a greater effect on the d_{33} than in the hard material. The error bars on the soft material can be seen to reduce as the pre load is increased, indicating that at very low pre loads the measurement is less reliable.

In many of the experiments using this kind of instrument it is difficult to separate the change in behaviour due to different experimental conditions and those due to the material itself. The highly piezoelectric PZT ceramics are notoriously dependant on factors such as stress level, temperature, time, and it can sometimes be more instructive to examine a less active but more stable material such as quartz. Figure 8 shows the variation of a quartz disc with increasing pre load, and the zero slope shows that the change that occurs with PZT ceramics is material and not instrumentation dependant.

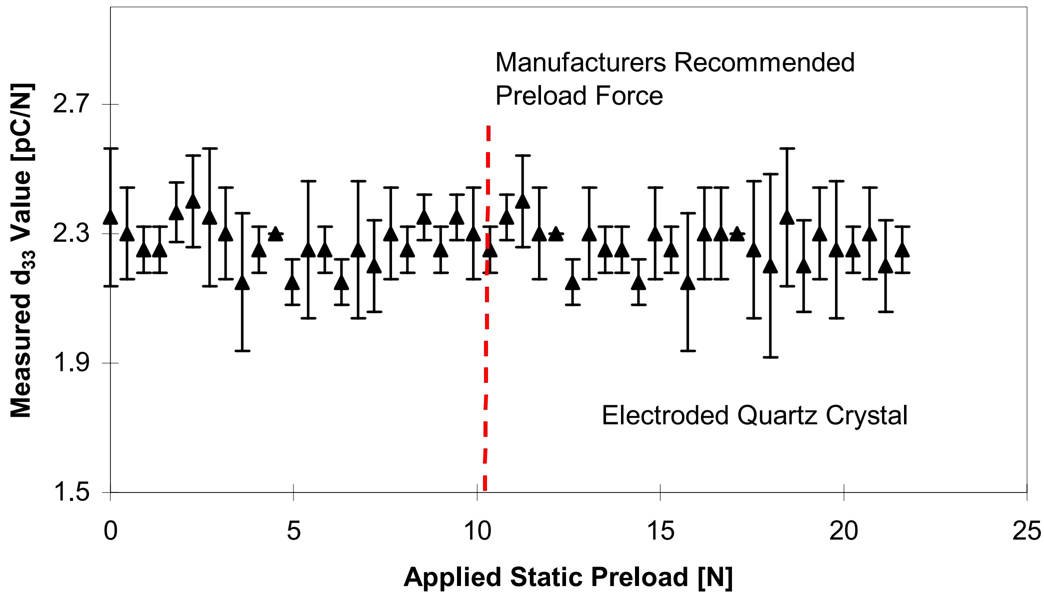

Figure 8: Effect of static pre load on Quartz Sample

3.4 Time Dependant Effects

In the investigation of the Berlincourt system it was found that there was a time dependent variation in the measurements. In all cases the measured d_{33} decreased with increasing measuring time under load. Figure 9 shows an example decay curve for a hard PC4D material

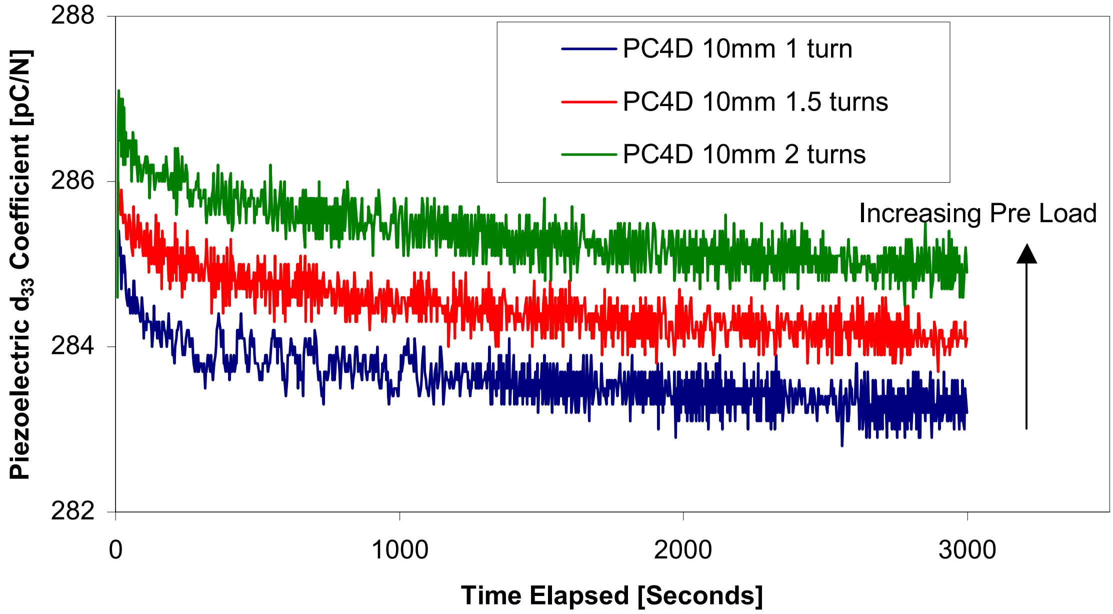

Figure 9: Effect of Static Pre load on relaxation of d_{33} on PC4D

at several different pre loads. Initially it was not clear if this was a measurement artefact dependant on the system. However, after a number of experiments it was proven to be related to the **static pre load** because of the following observations.

- If a sample is removed from the system and immediately replaced the behaviour continues along the same curve.
- Inserting a fresh sample at any point initiates a new decay/relaxation curve.
- If a sample is removed from the system and left to recover for 24 hours the relaxation curve begins again at time zero.
- If a sample is left in the system under pre load for several hours, but no AC measuring force then the relaxation curve is flat, i.e. most of the relaxation has already taken place.
- The behaviour is dependant on material, there is no measurable relaxation for lithium niobate.

The decay of d_{33} with time is difficult to measure, firstly because the changes are close to the resolution of the system and, secondly because the largest change occurs in the first few seconds and since it takes at least 5 seconds to get a stable reading the change is difficult to quantify.

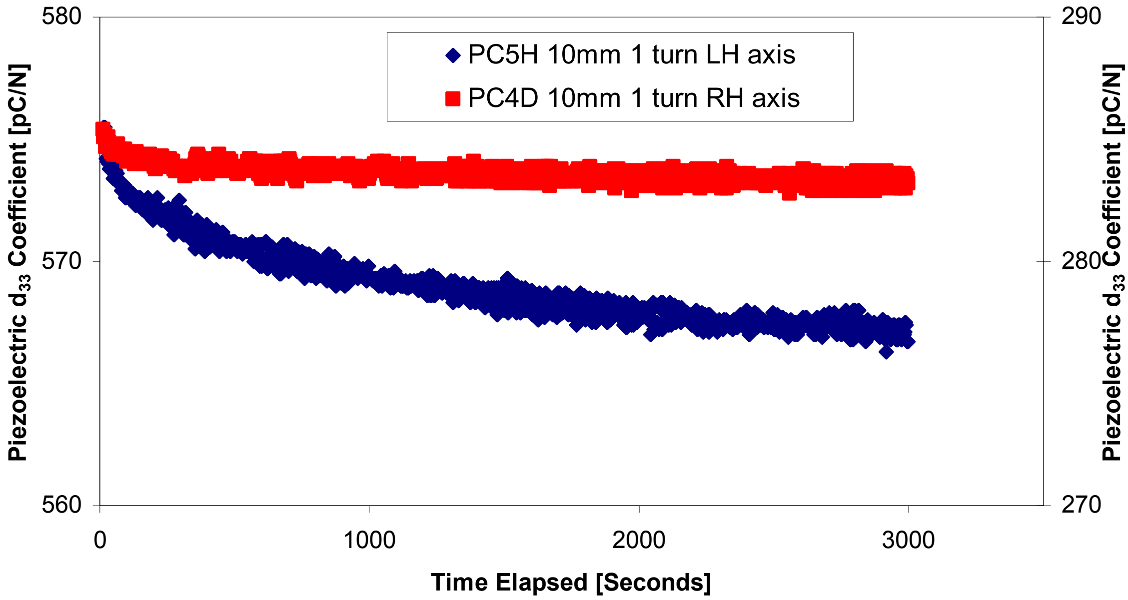

Figure 10: Relaxation of d_{33} in 10mm PC 4D and PC5H samples

The effect of static pre load on the decay in d_{33} is dependant on the material, Figure 10. For a given material the region where the decay flattens out is offset with the initial decay being similar. As seen with the static loading experiments, increasing the pre load increases the offset for a hard material, and decreases that for a soft composition. The different behaviour of hard and soft material is not just in the offset but also in the rate of relaxation. Figure 10 shows a comparison of a hard and soft sample, where the hard material is stable after 3500 seconds, yet the soft material is still relaxing. A general observation was that for equal experimental conditions it took longer for the soft samples to reach a stable region, although some of this could be attributed to the higher d_{33} of the soft material that may be measured more accurately for the same sensitivity.

Further evidence that the decay is due to some form of relaxation in the sample rather than instrumental artefact is seen in Figure 11, which shows the time decay curves for a thick and thin soft sample. The slope of the 1mm curve is much greater because of the much higher static stress concentration introduced in the thinner sample. An observation for the 1mm samples at various pre loads was that after an initial rapid decrease there was a slight rise at around 250 seconds before continuing the downward trend. Thus, maybe there are two competing mechanism for stress relaxation at the high stresses in the thinner samples.

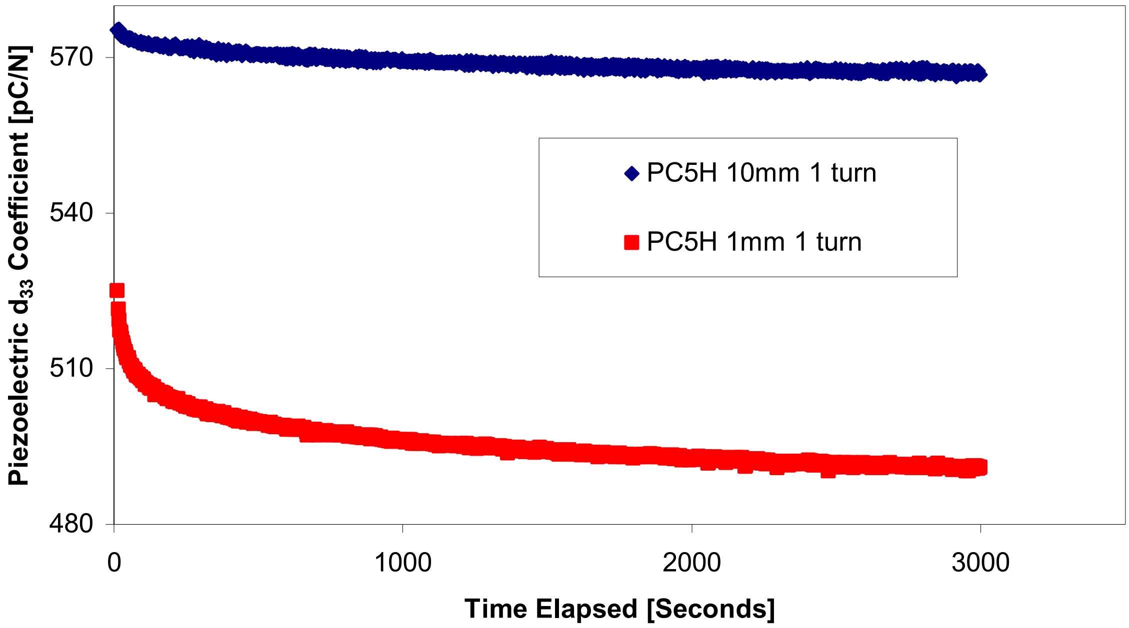

Figure 11: Comparison of d_{33} relaxation for a 10mm and 1mm PC5H sample

3.5 Calibration of System

In the Berlincourt system there are two calibration points, the zero calibration, and the gain setting. The zero calibration is simply that the reading for a non piezoelectric material should be zero, and the calibration/correction is achieved by placing a non conducting sample in the system and checking that the reading is zero.

The gain calibration issue is more difficult. For measurement instrumentation it is normal for the calibration of the system to be traceable to the national measurement system via the measurement of some electrical quantity or reference artefact. In the Berlincourt system the quantities measured are force and charge. The traceable calibration of the charge is relatively straightforward, but the calibration of force would be more difficult. Consequently in the Berlincourt system the force is measured by having a piece of PZT in the load train. This has the advantage that the charge signal from the internal reference can be compared to the sample charge to give a d_{33} measurement. This simplifies the measurement electronics, but does create a more difficult calibration problem. The solution is to use a reference artefact that has been calibrated by some other means, and to adjust the gain of the system to agree with the artefact. The normal reference artefact is a sample that has had the d_{33} measured by longitudinal resonance using the IEEE standard method. The disadvantage of this is that it is almost impossible to traceably calibrate this, and it also assumes that the value derived from resonance at around 100kHz is identical to that in the 30-300Hz range. This poses the question, '*at what frequency should the calibration be carried out?*'. We have seen from the discussion on the effect of frequency, that although the calibration may have been carried out at the 'standard' operating frequency, once away from this frequency the calibration route becomes more tenuous.

The material used in the reference material should obviously be stable with respect to time and temperature, and show little AC and DC load and frequency dependence. It should also have a high enough d_{33} to be able to calibrate the highest end of the measurement range. Quartz or Lithium Niobate would be ideal candidates but the d_{33} is too low to satisfactorily calibrate the higher ranges. Although the hard PZT compositions have roughly half the value of the most active soft materials, their improved stability with time make them a more suitable choice for a reference material.

Since, for independent measurements of d_{33} the reference sample will invariably be a thickness resonator, the thickness of the sample will be sufficient (probably at least 10mm long) so that the thickness effects seen previously will not be an issue.

3.6 Sample Geometry

An advantage of the Berlincourt method is that virtually any shape or size sample can be measured in the system, as long as there are electrodes to make electrical contact and apply the load. The method is supposed to be sample geometry independent, and in order to examine this a selection of samples from the same batch of material were examined.

Figure 12 a, b shows the effect of sample thickness on the measured d_{33} for a soft and a hard material, respectively. Over the entire range of thickness from 2mm to 10mm there does not appear to be a change in measured d_{33}. However, there is a definite difference between the 2mm and 1mm samples, with the soft PC5H material exhibiting a large decrease in the d_{33}, whereas the hard material showing a slight increase. Although the samples all come from the same batch of fired ceramic there could still be slight variations in the poling process for the individual thickness that could lead to differences in piezoelectric activity. In order to verify this, an experiment was devised where thin samples were stacked and compared with their thick counterparts. The results of these measurements are given in table 1. Assuming linear behaviour the measured d_{33} of a stack of thinner discs should be equal to the average of the

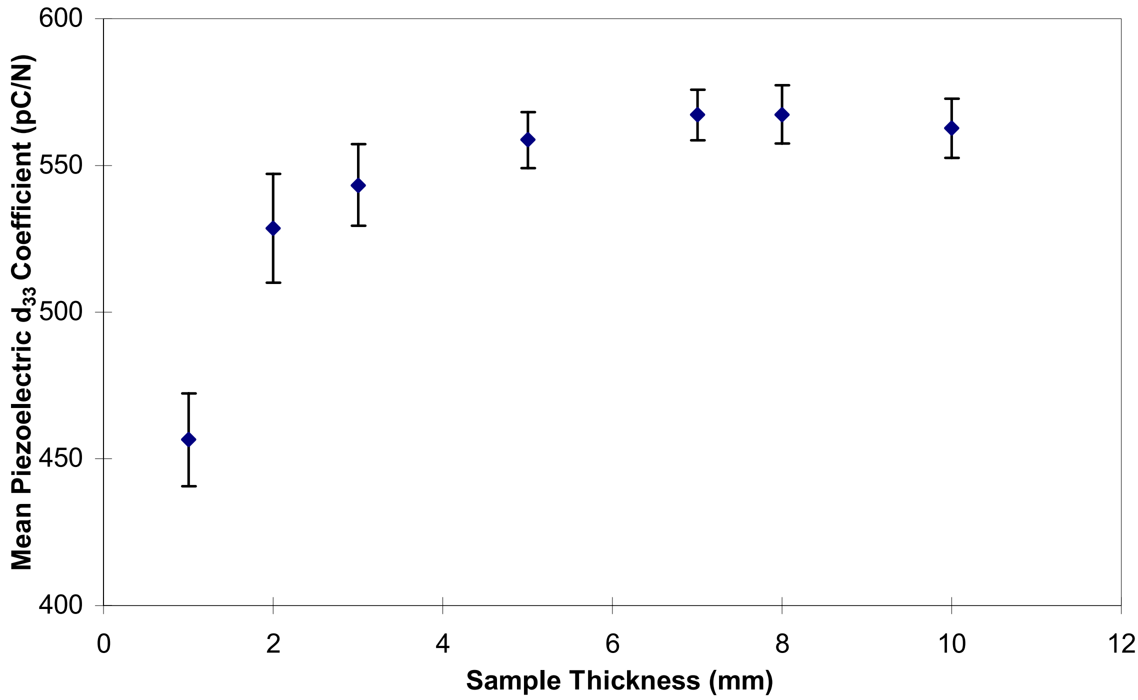

Figure 12a: d_{33} against thickness for a soft PC5H composition

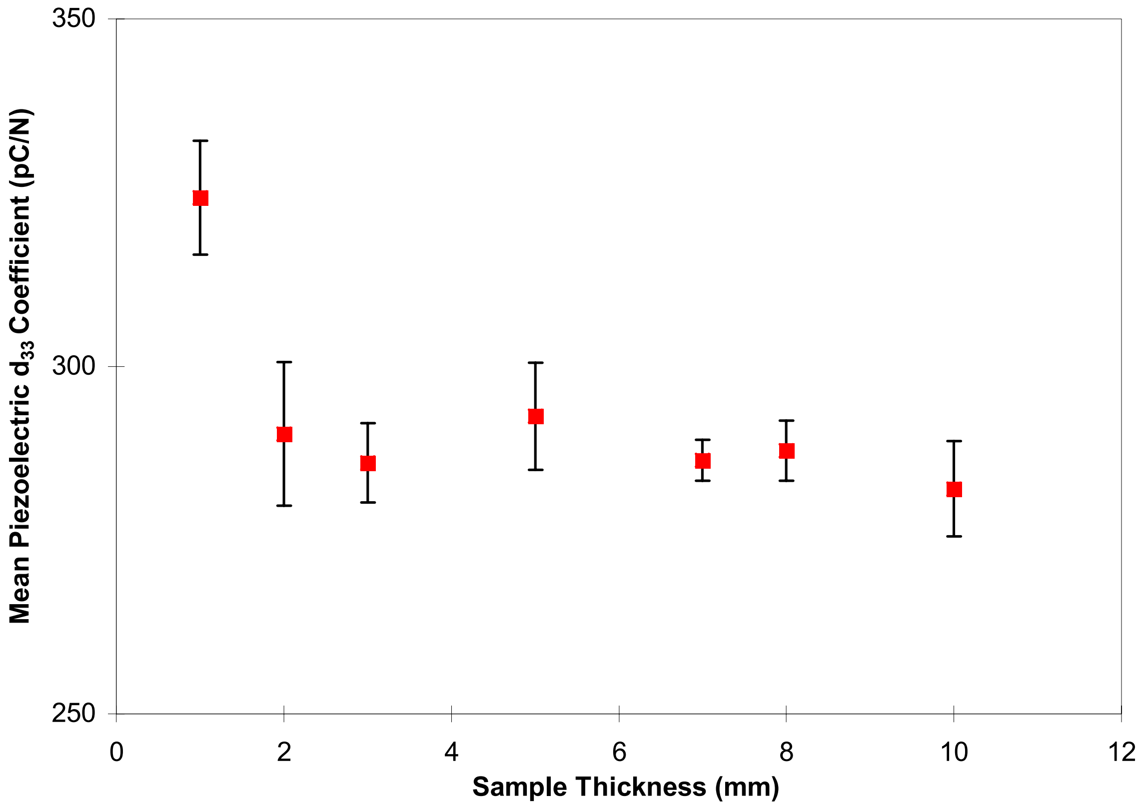

Figure 12b: d_{33} against thickness for a hard PC4D composition

individual discs. However this is not the case for the 1mm discs, and the d_{33} of two stacked 1mm samples tends towards the value for the 2mm samples. This indicates that the different thickness samples have identical piezoelectric activity, and the difference measured with the Berlincourt is *instrumental*.

Mean d_{33} Value [pC/N]		
Stack Height	Soft PC 5H	Hard PC 4D
1	456	324
2	555	316
3	577	302
4	559	288
5	577	293
6	565	297
7	571	296
8	572	292
9	562	291
10	568	289

Table 1 Effect on measured d_{33} of stacking 1mm discs

As discussed previously almost any shaped sample that can be inserted in the measuring head can be measured, however difficulties arise when the samples have a large area. For large samples it can be difficult to position the sample centrally such that the sample is balanced. If the sample is off centre it can introduce bending stresses that will affect the results. For very thin samples the misalignment of the loading pins can lead to considerable shear stresses, again giving misleading results.

For samples with a large surface area compared with the normal 10mm diameter a separate problem was identified, namely the neutralisation of the charge via the surface. It was noted that the reading on the meter display could be affected instantaneously by breathing over the sample, i.e. the extra moisture in the breath was leading to an increased charge leakage path. The value quickly regains the steady state value, but it is possible that convection currents could have a greater effect on larger surface area samples.

3.7 Loading geometry

The normal loading contacts for a Berlincourt system are hemispherical contacts of the order 4mm diameter. This point contact is the preferred mode because firstly the contact points for samples with rough surfaces is more controlled, and secondly it minimises any clamping in the lateral direction by the loading contact. Other types of contact test probes can be used and are more useful for more esoteric shaped samples. For a spherical sample or loading on the perimeter of a disc, a flatter loading probe will provide a more practical loading set-up and still maintain essentially point contact area. For very thin samples and films a flatter electrode will reduce shear due to misalignment, and also reduce the stress levels for a given measuring force.

The effect of the test probes on the measured d_{33} was investigated by using three type of test probes, the standard point contact, a flatter rounded contact (roughly 16mm diameter), and a completely flat probe with a diameter of 8mm. The contact area for the rounded probes was measured by examining the imprint of the area using carbon paper. For the standard contact the imprint was a circle roughly 0.5mm in diameter, and for the more rounded contact a diameter of around 1mm. The results for this experiment are shown in Figure 13 for a soft material with the standard pre load of 10N in each case. It appears that the increased contact area probes decrease the static stress, and so for the soft material the d_{33} increases whilst for the hard material the measured d_{33} decreases.

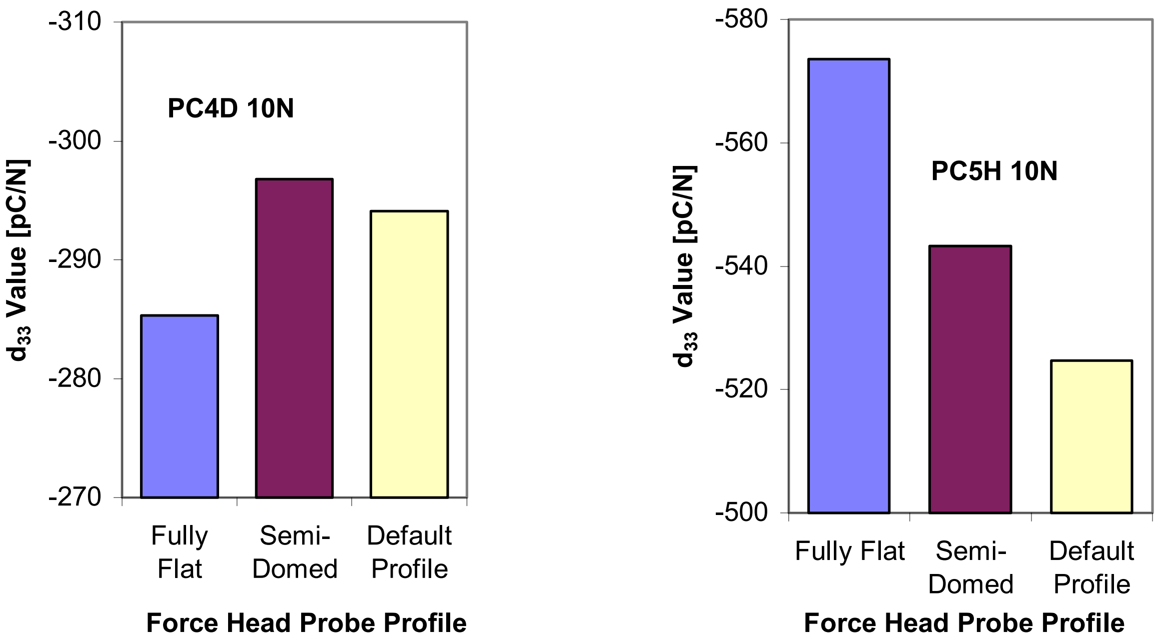

Figure 13: Effect of different loading probe on the measured d_{33}

3.8 Second Order Effects

Where it was practically possible the samples were left to recover between each individual measurement so that the change due to one particular experimental parameter could be observed. This was not always practical, for instance the sweep of d_{33} with frequency was performed with a sample continuously in the measurement head. An ideal test would be to measure at each frequency and remove the sample after each measurement and allow the sample time to relax. Another approach would be to leave the sample in the system for several hours before commencing the measurements. However, in the tests performed here there is a mixture of frequency and time dependant information. For instance if the frequency is swept first up to the high frequency and then back down again the behaviour is hysteretic. Figure 14 shows these sweeps for a soft material of 1mm thickness. The behaviour is hysteretic because the static load is applied at time zero and by the time the frequency sweep is finished almost an hour will have elapsed, thus showing the time dependent component of this experiment.

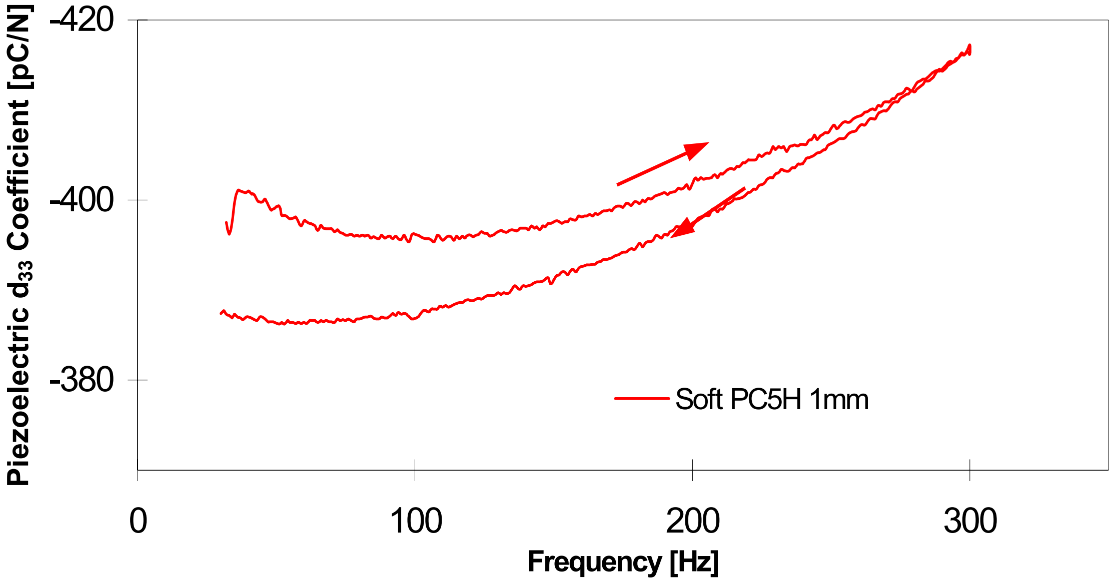

Figure 14: Up and down Frequency Sweep on PC5H 1mm thick sample

3.9 Differences between Hard and Soft materials under Static Load

In the most of the experiments discussed here there has been a trend for the hard material to behave in an opposite manner to the soft material. The most probable cause of this behaviour is due to the differing response of the materials to static loads applied parallel to the poling direction. For instance it is known that the permittivity for hard materials increases under applied stress, whereas the soft materials show a decrease. For the hard material this can be attributed to a de-aging, where any large mechanical or electrical excursion will initiate a new aging cycle. For the soft materials the decrease is due to the domain mobility effectively depoling the material under relatively low stresses.

This explains the difference in the static load behaviour seen in Figure 7. Also, considering the behaviour under different load contact probes it is apparent that the increased contact area probes decrease the static stress, and so for the soft material the d_{33} increases whilst for the hard material the measured d_{33} decreases. This could also be used to explain the thickness effect, where in very thin samples there are proportionately more regions under a high static stress so the static stress effect dominates. As the thickness of the sample is increased by stacking this high stress region is reduced, thus reverting back to the 'true' d_{33}.

4. Finite element and theoretical aspects of sample geometry/loading.

It is clear from some of the preceding experimental results, particularly the different response of various sample geometries that the Berlincourt test has some shortcomings. If the response of the piezoelectric is assumed to be linear then this predicts that the results should be independent of sample geometry or method of stress application.

If a finite element model of the measurement system is simulated using ANSYS finite element code, using linear coupled field elements, and assuming the loading state can be approximated as a static case, then the predicted d_{33} will be equal to the d_{33} used in the input. Although in the Berlincourt measurement a point load on a thin disc can give rise to triaxial stresses and stress concentrations, the linear assumption does not lead to any change in measured d_{33}. In the model each element acts as a linear stress-to-charge converter, and although the stresses may be unevenly distributed throughout the sample, the total charge is conserved.

There are few analytical solutions to the problem that might account for some of the variations seen experimentally. However, in the field of thin films there is an analytical solution to account for the clamping effect of a substrate. There are two solutions [Lefki and Dormans] to simulate the film d_{33} behaviour in the indirect and direct modes. In the direct mode (the application of a stress to produce a charge) the applied stress is transferred through the substrate to the film, and the film is thought to be held rigidly from moving in the 1 and 2 directions by the substrate. In the indirect mode (the application of a field to produce a displacement) the substrate is passive as the applied field does not induce a displacement, and the film is then clamped by an elastic substrate with a stiffness, *Y* and Poisson's ratio, σ

$$d_{33}(ip) = d_{33} - 2d_{31}\, s_{13}^E \big/ \left(s_{11}^E + s_{12}^E\right) \tag{6}$$

$$d_{33}(dp) = d_{33} - 2d_{31}\left(s_{13}^E + \sigma/Y\right) \big/ \left(s_{11}^E + s_{12}^E\right) \tag{7}$$

where equation 6 is for the indirect , and 7 for the direct mode.

For both cases the effect of the d_{31} term is to reduce the effective d_{33} since s_{13} is also negative (and of course d_{31} is negative). The boundary condition for the indirect case represents the extreme case and for the materials used in this work would mean a measured d_{33} of 0.33 times the true d_{33} for the PC5H and 0.45 for the PC4D. The magnitude of this clamping can be confirmed in the finite element model, by preventing the outside of the sample from moving. The reason why the behaviour is now apparently non linear is that a clamping force is needed to prevent the sample moving and this acts against the measurement force leading to a reduced charge output. Of course, the greatly reduced d_{33} predicted by equations 6 and 7 only applies to thin films on substrates, but similar boundary conditions could be envisaged in the Berlincourt experiment.

Consider the case where the force in the Berlincourt is spread out over the whole sample surface, and the friction is such that the sample surfaces are not allowed to move in the 1-2 directions. This is almost identical to the thin film analytical solution, except that the finite thickness allows some relaxation, giving a slightly higher d_{33} than predicted by equation 6. These boundary conditions do not precisely resemble the Berlincourt experiment, and consequently these large reductions in measured d_{33} are not seen.

If the area of load application is reduced then the clamped area reduces, and the clamping forces reduce until, when a point load is used, there is no clamping force and the measured d_{33} is once again the model input d_{33}. Figure 15 shows the change in measured d_{33} with changing the load area for a disc with a 10 to 1 radius to thickness ratio. The curves go from fully clamped where the d_{33} corresponds to equation 6 to the point load which gives the 'true' d_{33}. These curves change shape slightly for different radius to thickness ratios, but the end points are fixed. The stress distribution for a partially clamped sample, Figure 16, is fairly uniform under the loading region and the reduction in apparent d_{33} comes purely from the extra force needed to clamp the displacement at the loading interface.

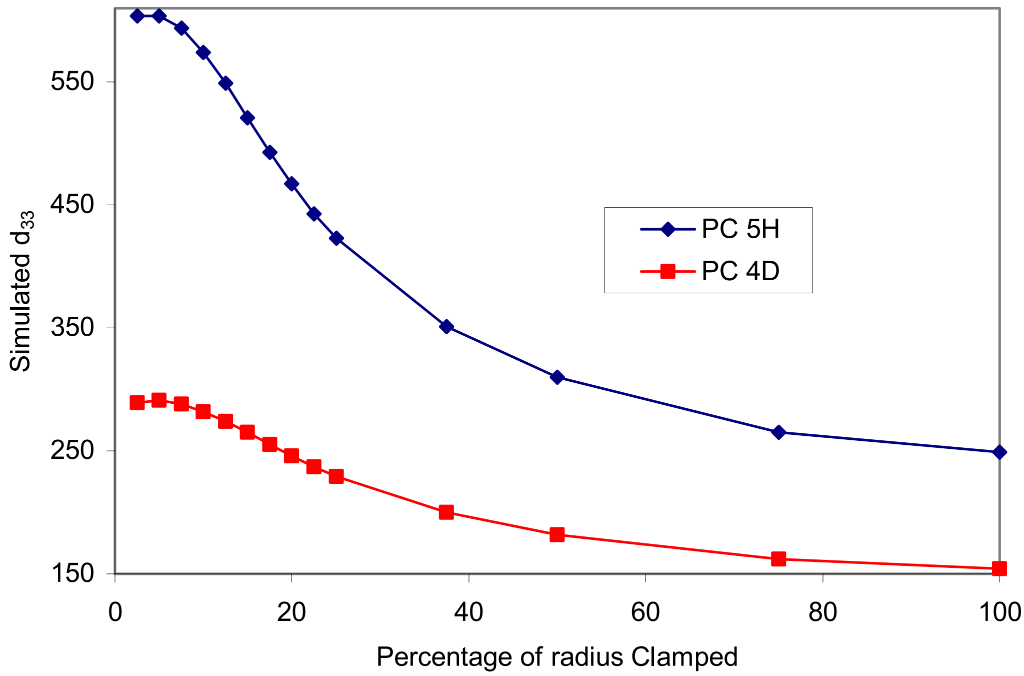

Figure 15: FEA simulation of direct mode experiment with increasing area of disc loaded and clamped. Disc radius to thickness ratio 10:1.

Another possible mechanism for introducing external boundary conditions could be due to the inertial resistance of the surrounding material under dynamic loading. This would effectively stop the outside of the sample from moving, and show up as a frequency dependence of the measured d_{33} response. However it is unlikely that this effect plays a major role at the frequencies used in the Berlincourt experiment.

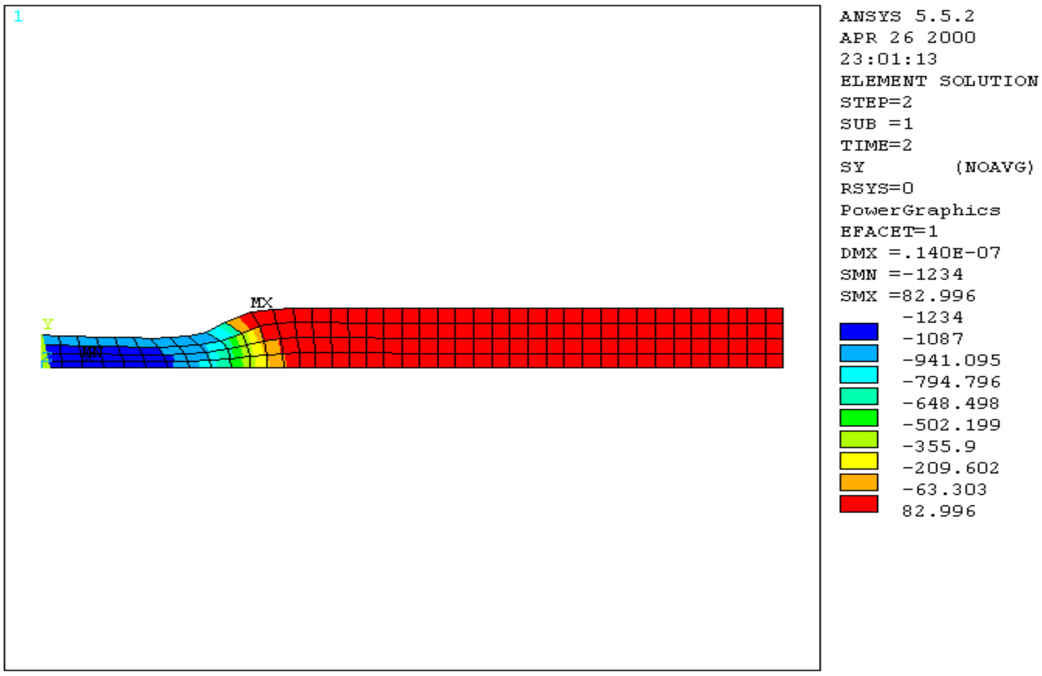

Figure 16: FEA simulation direct mode loading of 10:1 radius to thickness ratio disc, loaded and clamped over a quarter of the radius.

In all the preceding modelling of behaviour, the effect of clamping has been to reduce the measured d_{33}, however there are no plausible means by which clamping could increase the charge output, and thus the measured d_{33}.

To arrive at a simulation of the Berlincourt experiment where the measured d_{33} was increased some other theory is needed. In previous work [NPL Report CMMT(A)150] a methodology was developed to use ANSYS to model systems where the d_{33} was dependant on the stress level. Normally d_{33} is given as constant, but generally d_{33} is dependant on applied stress or field levels. In the previous work, the d_{33} was assumed to vary linearly with applied AC stress level, based on some independent experimental measurements. This can lead to either higher or lower measured d_{33} values depending on the variation of d_{33} with applied AC stress levels, and could be extended to deal with a d_{33} dependant on the level of DC stress.

4.1 Summary of Finite Element Modelling

In the Berlincourt test, if the material is linear then the measured d_{33} should be independent of sample shape, method of loading etc:

- unless there are experimental conditions which lead to clamping of the sample leading to a reduction in the load – for example
    - clamping in XY plane by the load applicator
    - dynamic response dependant (frequency dependant)

Otherwise the changes in response are because

- the d_{33} is intrinsically dependant on AC stress levels
- the d_{33} is intrinsically dependant on DC stress levels

5. Comparison of low field Berlincourt measurements with High Field/Stress

5.1 Measurement of d_{33} in the indirect mode

As previously mentioned, the main advantage of the Berlincourt method lies in its simplicity. However, for many piezoelectric applications, materials are used in the indirect mode-applying a field to achieve a displacement for actuation. This poses the question, *is the value of d_{33} determined by the Berlincourt method the same as that determined using the indirect method?*

For a discussion of how to perform d_{33} measurements of PZT materials using the indirect mode there is a good practice guide available [NPL Good Practice Guide No. 24].

Results for the indirect d_{33} measurement for a number of PC 5H samples are shown in Figure 17, where d_{33} is plotted against the applied electric field. In the linear theory of piezoelectricity d_{33} is a constant, i.e. it does not vary with the applied field or stress level. For materials such as lithium niobate this is a valid assumption, but for the soft PZT material measured here the d_{33} has doubled over the applied field range. This graph illustrates one of the fundamental differences between the direct and indirect measurement methods. In the indirect method because of the small displacements involved large fields are applied in order to produce more measurable displacements. The applied field/stress level is therefore a large AC field with zero DC level, and the material experiences tensile and compressive stresses. In contrast with the Berlincourt method the charge from a small AC stress can easily be measured, however a large DC stress bias is needed to hold the sample in place, so the sample

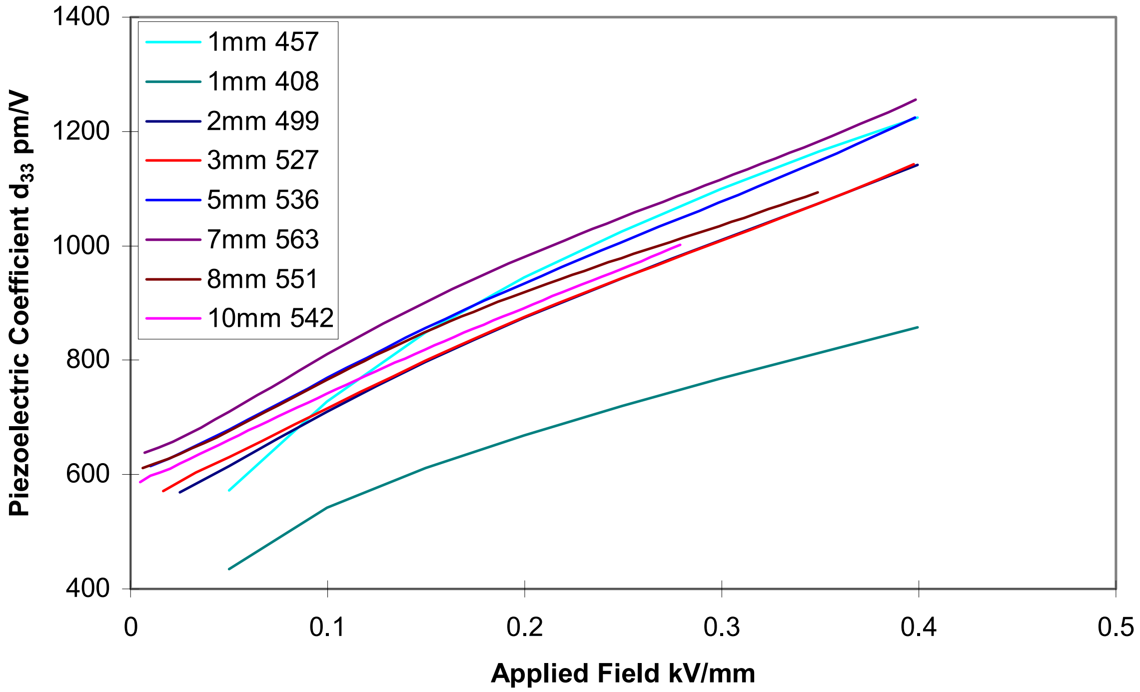

Figure 17: Indirect d_{33} measurements on PC 5H samples of varying thickness

is always in compression. This means that for a valid comparison between the measurements the displacement measurements should be extrapolated back to zero applied field levels, and the Berlincourt measurements need extrapolation back to zero pre stress levels.

Figure 18 compares the d_{33} indirect measurement at zero field with the normal Berlincourt measurement (i.e. a 10N pre stress). The extrapolation for the displacement measurements assumes that the behaviour is linear all the way to zero field. Considering Figure 17 this appears valid for the thicker samples, however the thinner samples show a tail off in behaviour as a field of 0.05kV/mm is approached. One problem with these thinner samples is that the actual displacements measured are smaller for equivalent fields. For the 0.5kV/mm field a d_{33} of 500 pC/N represents a displacement of only 25nm, and at these levels system noise and resolution is important. Comparing the two measurement results in Figure 18 the displacement measurement gives consistently higher results, by about 10%. However, the trend for the different samples is the same. Two 1mm samples were chosen for the first of the series of measurements in Figure 18 that gave different Berlincourt d_{33} values, and this is reflected in the displacement measurements. It is interesting to note that for a given composition the variations in d_{33} measured by the Berlincourt are reflected in the zero extrapolated displacement results, however the gradients of the displacement versus applied field measurements for d_{33} are similar for all the samples.

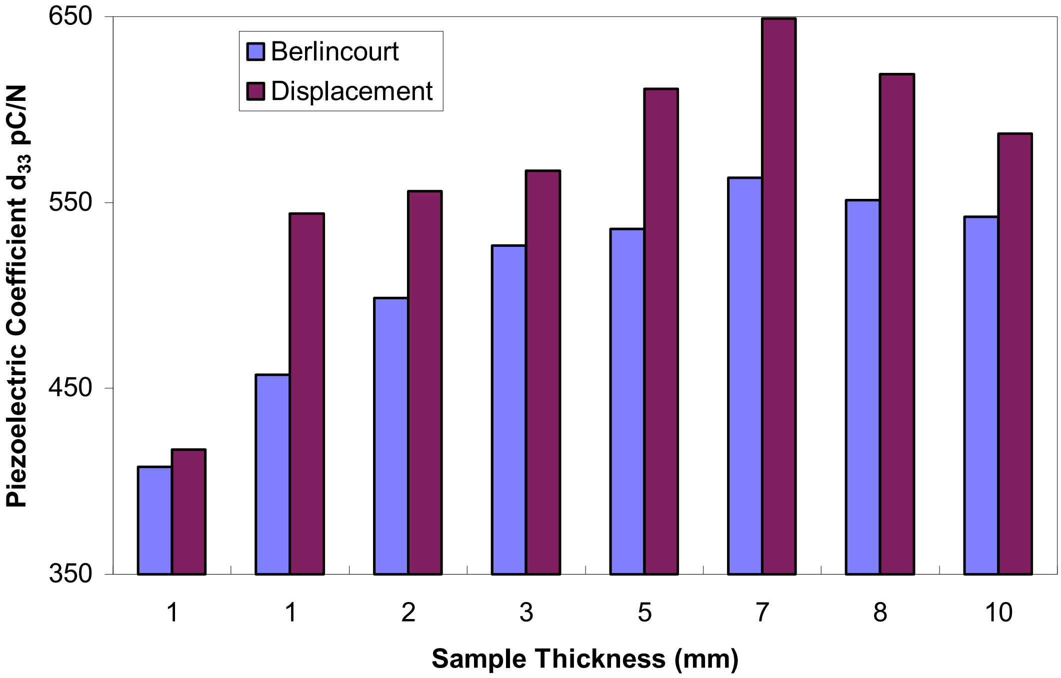

Figure 18: Comparison of indirect and direct measurements on PC 5H material

The disparity between the two measurement methods could be attributed to the fact that the Berlincourt measurements were taken at 10N pre stress. However, the pre stress should have a greater effect on the thinner samples, not a constant difference as observed. Similar measurements on the PC4D material also gave a higher displacement d_{33} than its corresponding Berlincourt value, but this time only by around 3%. If the difference was solely due to the pre stress then the difference should be reversed for the hard material. There are several possible interpretations as to why the measurements are different, including the following:

- Berlincourt measurements are done under a pre stress.
- The calibration of the Berlincourt is at a fixed frequency using a sample measured at resonance.
- In the displacement measurement method the sample experiences tension and compression whilst in the Berlincourt method it is always under compression.
- The boundary conditions for the two experiments are slightly different.

The discrepancies are not that large considering these issues, and it can be concluded that the Berlincourt results agree with the zero field extrapolated displacement measurements. However, the Berlincourt method gives no indication as to what might happen at higher driving fields.

5.2 Direct Measurements of d_{33} at High Mechanical Stress Levels

In the previous section we have seen that there is equivalence between the Berlincourt measurements and the displacement measurement results when extrapolated back to zero applied field. However, the Berlincourt data cannot be used to predict the displacement of materials at the kinds of fields used in practice. Obviously the Berlincourt result gives us the base level, and some kind of generic rate of change of d_{33} with applied field could be used to give an improved estimate. It may be possible to get an idea of how much the d_{33} changes with increased *field* levels by using increased *stress* levels, by using the equivalence of the indirect and direct piezoelectric behaviour. There are two experimental problems with this approach. Firstly for equivalence with the applied AC field experiments the Berlincourt measurement system would need to generate *tensile* and compressive stresses. The difficulty with applying tensile stresses could be overcome by comparing with field-strain measurements performed with a DC bias to keep the stresses compressive. The second problem is that using the conventional Berlincourt system with a voice coil drive it is difficult to generate large stresses at high frequency. This second problem can be overcome by using a piezoelectric actuator to produce the AC stress.

The results of making direct piezoelectric coefficient measurements at high applied AC stress is shown in Figure 19. Clearly the behaviour is similar to the displacement behaviour under high applied fields, i.e. there is a near linear increase in d_{33} with applied stress, suggesting this is a possible method for predicting high field displacement behaviour.

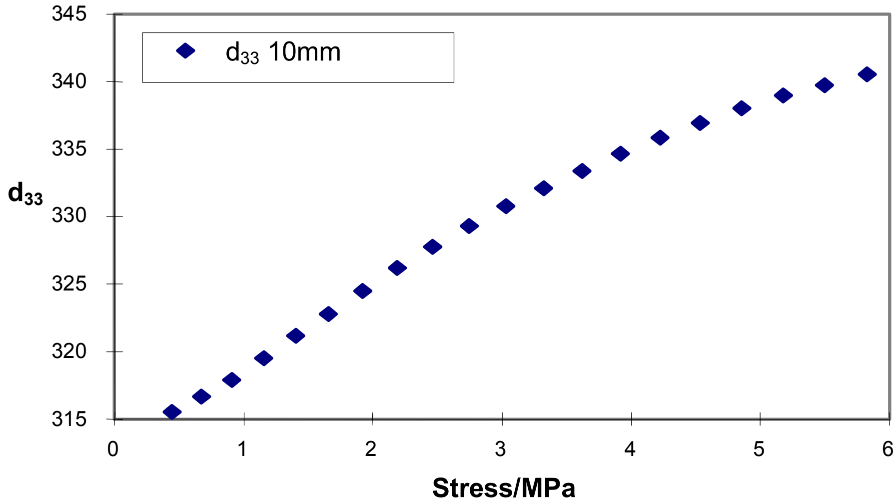

Figure 19: Piezoelectric coefficient, d_{33} with amplitude (pk/pk) of ac pressure

6. Acknowledgements

Thanks are due to the industrial sponsors of this work. Many of the measurements made in support of this good practice guide were carried out using a piezometer supplied by David Glasspoole of Take Control, on PZT samples from Howard Thomas at Morgan Electroceramics, and Mark Henson of Advanced Ceramics.

7. Annex

7.1 Commercially Available Equipment

Detailed in the following section is a selection of instrument manufacturers currently producing d_{33} measurement apparatus based around the direct effect quasi-static measurement schema. The National Physical Laboratory does not offer any recommendation for the equipment produced by these companies, nor does it guarantee the validity of the information supplied with respect to the most current instrument specifications.

7.1.1 TakeControl Piezometer Systems

Models PM15, PM25, PM35

Institute of Research & Development
University of Birmingham Research Park
Vincent Drive
Birmingham
B15 2SQ
United Kingdom
Tel: +44 (0) 121 415 4155
Fax: +44 (0) 121 415 4156
http://www.take-control.demon.co.uk

7.1.2 Sensor Technology Limited

Piezo-d Meter

P.O. Box 97, 20 Stewart Rd.,
Collingwood, Ont., Canada
L9Y 3Z4
Tel: (705) 444-1440
Fax: (705) 444-6787
e-mail: techsupport@sensortech.ca
http://www.sensortech.ca/

7.1.3 American Piezo Ceramics International Ltd [APC]

Pennebaker Model 8000 Piezo d_{33} Tester

PO Box 180, Duck
Run Mackeyville, PA 17750 USA
Tel: +570-726-6961
Fax: +570-726-7466
e-mail: apcltd@cub.kcnet.org
http://www.americanpiezo.com/

7.1.4 Channel Industries, Inc

839 Ward Drive
Santa Barbara, CA 93111 USA
Tel: +(805) 967-0171
Fax: +(805) 683-3420
e-mail: ciisales@channeltech.com
http://www.piezoceramic.com/

7.2 Sample Materials Specifications

The piezoelectric ceramic materials investigated in this work were supplied by Morgan Electroceramics - Unilator Division. The two materials examined were both Lead Zirconate Titanate - with the hard material designated PC4D and the soft material designated PC5H. The PC4D is a Navy type I composition with a low loss, designed for use in high power transducers, such as sonar transducers and ultrasonic cleaning. PC5H material is a Navy

			Matroc Unilator Materials	
Parameter	**Symbol**	**Unit**	**PC4D**	**PC5H**
Electrical - Low Field				
Relative Permittivity	ε^t_{r33}		1325	3200
Dielectric Loss	tan δ		0.002	0.018
Resistivity		Ωm	10^{10}	-
Electro-Mechanical				
Coupling Factors	k_p		0.58	0.6
	k_{31}		0.32	0.35
	k_{33}		0.47	0.72
	k_t		0.45	0.5
Charge Constants	d_{31}	x 10^{-12} C/N	-123	-250
or Strain Constants	d_{33}	x 10^{-12} C/N	335	620
	d_h	x 10^{-12} C/N	89	125
Voltage Constants	g_{31}	x 10^{-3} Vm/N	-10.5	-8.7
or Stress Constants	g_{33}	x 10^{-3} Vm/N	28.6	21.9
	g_h	x 10^{-3} Vm/N	7.6	4.4
Quality Factor	Q_m		600	65
Density	ρ	kg.m^{-2}	7650	7400
Curie Temperature	T_c	°C	330	200

Abstract from Morgan Matroc - Unilator Division Materials Data Sheet

type VI material, with a high piezoelectric coefficient, making it useful for high sensitivity receivers and high displacement actuators.

7.3 Round Robin tests

As part of the measurements carried for this good practice a small round robin was undertaken to confirm some of the findings. Two laboratories using similar instrumentation measured a set of hard (PC 4D) and soft (PC 5H) samples 10mm diameter, and thicknesses of 10mm and 1mm. The measurements were all performed at the same frequency, under different levels of pre load. The results of the round robin confirmed

- The variability of the soft material is greater than the hard.
- The variability of thin samples is greater than for thick.
- The hard material is less dependent on the pre load.
- The d_{33} of the soft material decreases with increasing pre load.
- The d_{33} of the hard material increases with increasing pre load.
- The difference in d_{33} for thick and thin materials is consistent with the behaviour under the application of a pre load.
- Thick samples are less affected by the pre load.

The spread of results in this round robin has also been reduced as a result of controlling the conditions under which the measurements were performed. Figure 20 shows the results for the current round robin compared with a study on a similar material. The data has been plotted as a percentage deviation from the average for each laboratory, and clearly shows the reduced scatter from close control of the experimental set up. Clearly the control has been achieved by using almost identical piezometers, and further work is needed to maintain this using different makes and models of direct testing machines.

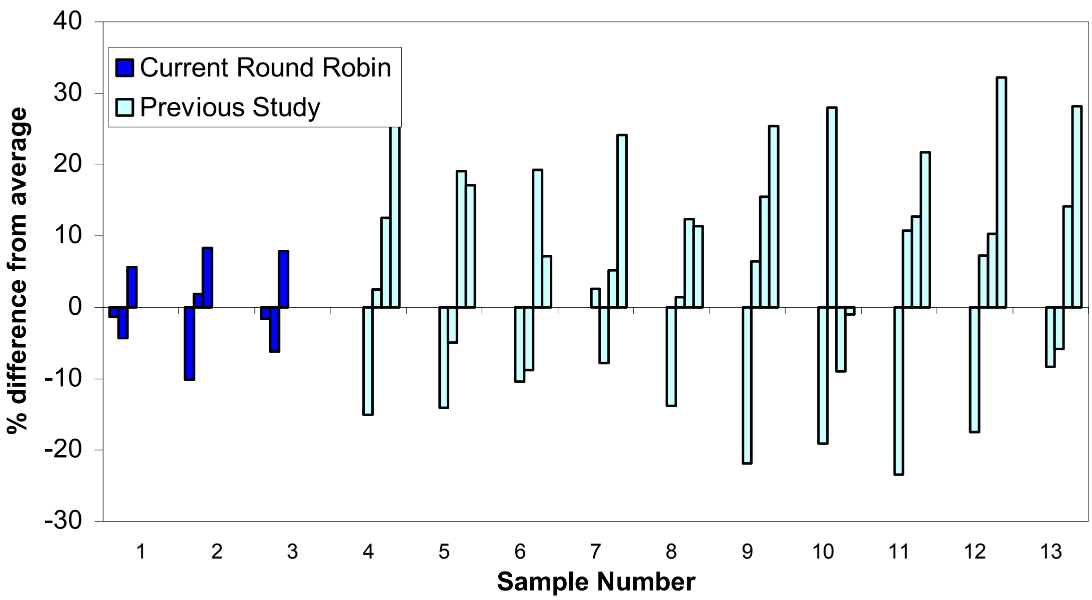

Figure 20: Round Robin tests using Berlincourt measurements on PC 5H material

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Standards

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