

**6MPI0301:
Losses in Piezoelectric
Materials**

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6MPI0301: Losses in Piezoelectric Materials**Markys Cain, Maria Lodeiro, Mark Stewart and Matthew Theobalds**

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ABSTRACT

The losses associated with piezoelectric behaviour have been investigated, by measurements at frequencies and fields where extrinsic contributions play a significant role. The study concentrated on measurements in the quasi static frequency range 0.1 to 1kHz, rather than through resonance methods. The piezoelectric loss was measured using a standard Berlincourt type setup modified to enable the phase angle between the applied stress and charge output to be analysed. The mechanical loss was also investigated by measuring the phase angle between applied stress, and induced strain in a strain gauged piezoceramic sample.

The piezoelectric loss showed an increase with increasing AC amplitude for hard and soft. However, the increase in loss seen at low frequencies was seen in all materials including Lithium Niobate, and it was concluded that this was an experimental artefact. The elastic loss measurements were more difficult to quantify because of bending of the sample, giving rise to arbitrary initial phase angles.

Introduction

Functional materials, including piezoelectric, ferroelectric, electrostrictive and others, are used in more and more demanding applications. This is due to the drive towards automation (sensors everywhere), miniaturisation (MEMS technologies) and autonomous resource-sparing philosophies (smart systems and structures). The deployment of functional materials in many industries and applications is limited as the various barriers of measurement, design, materials selection and economics associated with such hostile environments are being met. The increased demand for greater functional output has led to operation at higher power levels and this in turn has led to problems associated with energy loss in the system. These problems can range from simply requiring more power input, or unacceptable sound generation, to complete device failure through thermal runaway. Some applications may not be economically viable because they require expensive thermal cooling of devices. Designers facing these issues would benefit from an ability to predict the effects of loss by modelling behaviour. In order to achieve this, the mechanisms of loss in these materials must be understood, quantified and predictive models developed.

This project had aims to investigate new measurement methods that can characterise the losses associated with typical industrial piezoelectric ceramic materials. This functional interim project has also provided continuity between the completed CPM and the new MPP projects. The Rutherford Appleton Laboratory neutron diffraction beam-line time (£12,000 contribution) won by the project leader this summer (2002) was unfortunately postponed due to beam line problems at the ISIS facility, in Oxfordshire. The experiments will be performed later in the next beam-line schedule. These experiments will aid in the understanding of the mechanisms prevalent in PZT polycrystalline ceramic materials under high stresses and electric fields.

LOSSES IN PIEZOELECTRIC MATERIALS

A piezoelectric material can behave as a dielectric, as an elastic material, or a piezoelectric, depending on the boundary conditions imposed on the material. It can behave like a capacitor dielectric, moving charge in response to an electric field. Unless the material is an ideal dielectric there will be a power loss associated with this charge movement, which is termed the dielectric loss. This power loss is evident in the phase lag between applied field and dielectric displacement, and hence the loss is measured as a phase angle, termed δ . The power loss in a capacitor can be expressed simply by

$$\text{Power loss} = \omega CV^2 \tan \delta$$

where C is the capacitance, V the voltage, and ω the angular frequency.

A piezoelectric can also behave like a passive elastic material, where following Hookes law an applied stress will cause the material to deform. Again unless the material is perfectly elastic there will be an energy loss associated, giving rise to sound and heat, and this is termed the mechanical loss. Lastly the material is also piezoelectric, so an applied electric field will also induce a strain, and an applied stress will lead to a movement of charge. The phase lag between stress and charge, or field and strain is termed the piezoelectric loss.

These energy losses can result in sample heating in ultrasonic applications or noise production and hysteresis in actuators. This is why for many applications understanding the loss mechanisms and their absolute values becomes important [1].

COMPLEX REPRESENTATION OF LOSS

For a simple dielectric material the current voltage relationship for the application of sinusoidal voltages can be represented in terms of complex quantities. Thus the permittivity becomes

$$\epsilon_r^* = \epsilon_r' - j\epsilon_r''$$

Where ϵ_r' and ϵ_r'' are the real and imaginary parts of the complex relative permittivity. These quantities can be related back to the phase angle description of loss by

$$\tan \delta = \frac{\epsilon_r''}{\epsilon_r'}$$

For the description of losses in piezoelectric material this is a useful methodology, whereby the imaginary part represents the lossy part. Holland [2] was one of the first workers to rigorously analyse the mathematics detailing the concepts of complex coefficients to represent electrical, mechanical and piezoelectric losses. He showed that a positive loss corresponds to a negative loss in the phase angle. It was also shown that for any coefficient it was possible to have either positive or negative loss, however the overall power loss must of course must not be non-negative. In applying the formulation to a poled ferroelectric, various geometric and mathematical constraints give rise to the following inequalities for the imaginary components

$$\begin{aligned} s_{11}^{E''}, s_{33}^{E''}, s_{44}^{E''}, s_{66}^{E''}, \epsilon_{11}^{X''}, \epsilon_{33}^{X''} &\geq 0 \\ s_{11}^{E''} &\geq |s_{12}^{E''}| \\ s_{11}^{E''} s_{33}^{E''} &\geq (s_{13}^{E''})^2 \\ s_{11}^{E''} \epsilon_{33}^{X''} &\geq (d_{31}^{''})^2 \\ s_{33}^{E''} \epsilon_{33}^{X''} &\geq (d_{33}^{''})^2 \\ s_{44}^{E''} \epsilon_{11}^{X''} &\geq (d_{24}^{''})^2 \\ s_{33}^{E''} (s_{11}^{E''} + s_{12}^{E''}) &\geq 2 (s_{13}^{E''})^2 \\ \epsilon_{33}^{X''} (s_{11}^{E''} + s_{12}^{E''}) &\geq 2 (d_{31}^{''})^2 \end{aligned}$$

Interestingly these inequalities imply that if a material is either perfectly elastic or dielectric, then it will also be perfectly piezoelectric, i.e. the piezoelectric loss is zero.

Uchino and Hirose [3] have extended the complex representation of the mechanical, electrical and piezoelectric behaviour, and used the term “intensive” losses, where intensive means externally controllable physical parameters such as stress and field. The complex representation for the *intensive* mechanical, electrical and piezoelectric losses are

$$\begin{aligned} s^{E*} &= s^E (1 - j \tan \phi') \\ \epsilon^{X*} &= \epsilon^X (1 - j \tan \delta') \\ d^* &= d (1 - j \tan \theta') \end{aligned}$$

and the phase angles are respectively,

- ϕ' the phase delay between stress and strain under constant electric field E, (short circuit)
- δ' the phase delay between electric displacement and electric field under a constant stress X (zero stress, usually measured at 1kHz)*
- θ' the phase delay between strain and electric field at zero stress or the phase delay between charge and stress at constant electric field (short circuit).

The complex description of the *extensive* mechanical, electrical and piezoelectric losses, the materials internal losses, are

$$\begin{aligned} s^{D*} &= s^D (1 - j \tan \phi) \\ \epsilon^{x*} &= \epsilon^x (1 - j \tan \delta) \\ h^* &= h (1 - j \tan \theta) \end{aligned}$$

and the phase angles represent,

- ϕ the phase delay between stress and strain under constant electric displacement D, (open circuit)
- δ the phase delay between electric displacement and electric field under a constant strain x, (zero strain, completely clamped, usually measured far above resonance)*
- θ the phase delay between open circuit field and applied stress or the phase delay between applied charge and strain developed.

* the measurement of dielectric loss at 1kHz is an industry standard for capacitors, where the boundary conditions are not important, and usually termed $\tan \delta$. In this text this $\tan \delta$ is termed $\tan \delta'$ for consistency with the previous papers by Uchino[3].

Ikeda [4] has developed a number of relationships between the intensive and extensive losses, based on the coupling coefficient k. The losses are coupled, so it is difficult to isolate individual behaviour. For instance the intensive piezoelectric loss includes the intensive dielectric and extensive mechanical and piezoelectric loss as follows

$$\theta' = \phi' + \delta - \theta$$

HOW IS LOSS MEASURED?

As described previously, the loss is the phase angle between two mesaurands, and this angle can be measured using either a gain phase analyser, or lock in amplifier with a suitable accuracy and resolution. For some measurements this is simple, such as the measurement of dielectric loss where the measurement of current and voltage is relatively straightforward. For some of the other measurements the boundary conditions may not be easily realised experimentally, for instance it is difficult to completely clamp a piezoelectric to give zero strain for the measurement of δ . In fact rather than physically clamp the material, the measurement is carried out at very high frequencies where the displacement is so small that it is assumed to be zero strain.

In the study of vibrations and resonating bodies the quantity Q_m , related to the damping, is defined by the sharpness of the resonance. Q_m is the mechanical quality factor and is the inverse of the mechanical loss and can be estimated by the width at $1/\sqrt{2}$ of the maximum. As piezoelectric materials can be made into resonating devices this is a simple method of obtaining the mechanical loss. Of course the fact that the sample is resonating implies for these materials that the measurements are made at frequencies in the 10's of kHz and above, when the loss at lower frequencies may be different.

An extension to the simple method of analysing mechanical loss from resonant devices has been developed by many workers [2,5,6]. Resonance is routinely used to determine the fundamental materials parameters, using the resonator dimensions, a knowledge of the resonance mode, and the resonant and anti resonant frequencies [7]. Sherrit et al [6] have extended this methodology, using the complex description of materials parameters to model the whole of the resonant curve, i.e. the resonant peak widths as well as the frequency. This is presently the only method by which all the complex coefficients can be determined using one experimental method.

Another method of determining the losses in piezoelectric materials derives from the measurement of polarisation field hysteresis behaviour, commonly called PE loops. Here the polarisation of the material is measured whilst applying a sinusoidal field, and the result plotted on an X-Y graph. This measurement is fundamental for the investigation of ferroelectric behaviour, and the shape of the curves is indicative of the material type, hard or soft. The area within the PE loop, the hysteresis, is a measure of the loss and can be quantified as follows.

The relationship between polarisation and field can be represented by complex quantities as follows

$$P^* = \epsilon_0 \epsilon_r^* E^*$$

and the magnitude of ϵ_r^* is found from the amplitude of the polarisation and the electric field waveforms from,

$$|\epsilon_r^*| = \frac{|P^*|}{\epsilon_0 |E^*|}$$

The area within the PE loop is the energy loss per cycle and is similar to the expression for the power loss in a capacitor,

$$\text{Area within loop} = \pi \epsilon_0 \epsilon_r'' E_0^2$$

The area within the loop can be determined by numerical integration of the PE loop, and the real part of the permittivity can be calculated from the following

$$\epsilon_r' = \sqrt{(|\epsilon_r^*|)^2 - (\epsilon_r'')^2}$$

and the loss is simply the imaginary part divided by the real.

A similar procedure can be followed for all the other mesurands to determine the relevant losses, for instance stress strain will give the mechanical loss.

Various authors have highlighted their concerns over the definition and realisation of the 'piezoelectric loss'. In ways similar to the interpretation of mechanical and dielectric loss, the

piezoelectric loss has been described as the hysteresis between stress-charge density or the phase angle between charge and stress (or strain and field). However, close inspection of the units for this calculation show that the product is not an energy. Nevertheless, it is understood in the literature that the phase angle between charge and stress is indicative or related to energy loss or energy gain and we shall also use the term in this manner.

MECHANISMS OF LOSS IN PIEZOELECTRIC MATERIALS

So far, losses in piezoelectric materials have been described without considering how these losses come about. Obviously in order to be able to model the behaviour it is necessary to understand the mechanisms and the factors that control them at the microstructural scale. The experiments in this work will not be able to probe the behaviour at this level, however by studying different materials and conditions it may be possible to determine the most likely mechanisms.

The losses in ferroelectric ceramics have been ascribed to originate from four different sources [8]:

1. domain wall (internal microstructural features) motion against forces associated with imperfections, internal electrical fields, dopant and vacancy interactions etc.
2. intrinsic (fundamental) lattice dynamics
3. microstructural defects such as grain boundaries, microcracks etc
4. d.c conductivity in highly ohmic samples

In this report the focus is on losses at high fields and stresses, at frequencies below 1kHz. Here the dominant mechanism for loss is expected to be from internal domain wall motion (extrinsic piezoelectric contribution).

An early attempt to describe the loss in ferroelectrics was provided by Lewis [9], where the dielectric (and mechanical) loss was attributed to the movement of domain boundaries under the action of applied electrical or mechanical stress. The concepts of domain ‘pinning’ were also alluded to and the various contributions to the loss from the intrinsic (dipole) and ‘extrinsic’ (referred to as micro and macro-hysteretic) states were discussed. Some of the first observations of 90° domain walls were made in barium titanate single crystal materials which led to characteristic equations of motion incorporating viscous, inertial and domain pinning terms [10]. More recently, the concepts of modelling and measuring these separate losses present within piezoelectric materials have been developed [11,12,13] using viscous type descriptions of domain wall motion, vibration and oscillation. These efforts did not include the experimentally observed field dependence of piezoelectric coefficient and loss, which was investigated by considering contributions to the piezoelectric response by extrinsic domain wall vibration by invoking an adaptation of the Rayleigh model for ferromagnets [14,15]. This model described the hysteresis observed in sub-switching fields between electrical charge and applied mechanical stress, and has its basis in the formal Preisach theories. Such ideas are based on hysteretic systems that are described by a collection of bi-stable units with well-defined and distributed characteristics (coercive field, bias etc.) [16]. Such a description has recently been shown to offer a real world view of ferromagnetic behaviour embodied in a random walk motion of domain walls within a pinning field [17].

EXPERIMENTAL PLAN

As discussed the purpose of this study is to investigate the losses associated with piezoelectric behaviour, by measurements at frequencies and fields where extrinsic contributions play a significant role. Measurements of mechanical and piezoelectric loss are usually carried out at resonance frequencies, because it is easy to measure the mechanical loss by means of the resonant peak width. However this is usually in the 100's of kilohertz range where many of the loss mechanisms prevalent at low frequencies may not be in operation. This study therefore concentrates on measurements in the frequency range 0.1 to 1kHz. The measurement of dielectric loss at quasi static frequencies is a relatively trivial exercise and was not studied extensively in this study.

The piezoelectric loss was measured using a standard Berlincourt type setup modified to enable the phase angle between the applied stress and charge output to be analysed. The mechanical loss was also measured by analysing the phase angle between applied stress, (measured with a load cell), and induced strain (measured using a strain gauge attached to the sample).

MATERIALS

Single crystal quartz, as used in many quartz load cells, is a piezoelectric, but not ferroelectric, so it does not exhibit the hysteresis seen in ferroelectrics, and so can be regarded as having zero piezoelectric loss. The piezoelectric behaviour is also unaffected by frequency and increased stress or electric field levels, and so is a good choice for a standard material to check the calibration of the piezoelectric loss experiments. Unfortunately the piezoelectric output of quartz is over 100 times lower than the output from conventional PZT ceramics, and so is difficult to measure in them same experimental system without a change in the gain of the system, which could affect the calibration. To overcome this problem, single crystal Lithium Niobate, which has approximately 10 times the output of quartz, and is also similarly stable with respect to frequency and stress, was used to examine the performance of the system.

In this study, we aimed to further study the measurement of loss and especially the differences expected between soft and hard materials. The dispersive relationship exhibited by soft materials has been linked to its intrinsic domain dynamics under modest electric field. Soft materials have a low coercive field, high losses and high piezoelectric activity. The piezoelectric effect is dominated in soft materials by their large extrinsic contribution from piezoelectric domain motion observed under the application of electrical fields. In contrast hard materials show a higher coercive field, lower hysteretic losses and typically lower piezoelectric response. Hard materials do not generally exhibit domain motion until much higher electrical fields.

The PZT materials used were typical commercial compositions, a hard type PZT 4D, most often used in high power applications such as ultrasonic cleaning, where low loss is required. A very soft PZT 5H composition, which is used where large output is required but not for a high duty cycle, for instance quasi static actuators. Lastly a soft PZT 5A composition was used in the mechanical measurements, mainly because samples in the required size were available. The PZT 5A is similar soft composition to the 5H, however some of the high piezoelectric output has been 'sacrificed' by an increased Curie temperature, which enables higher temperature operation.

Measurement of Piezoelectric Loss

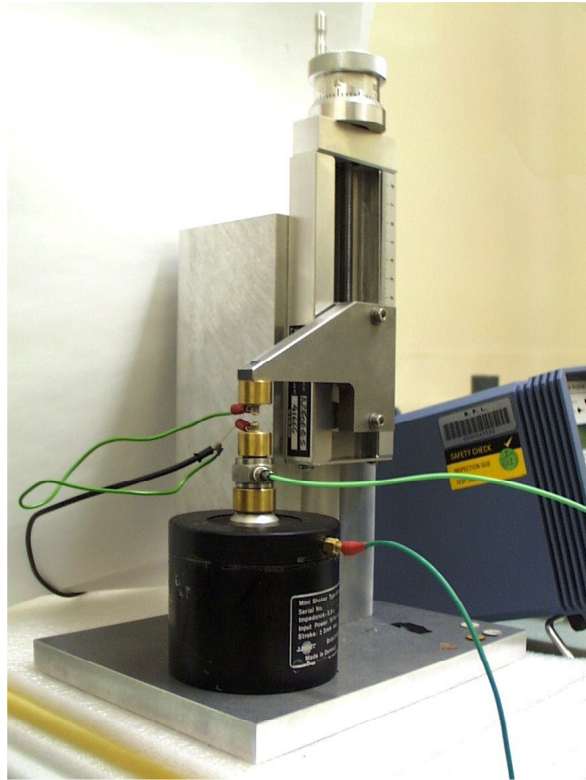


Figure 1 – NPL piezoelectric coefficient meter.

A simple piezoelectric coefficient meter, based on the direct Berlincourt-type method, shown in Figure 1, was designed and built. The sample is placed between two hemispherical contacts of diameter 4mm, under a small static pre-load, and an alternating mechanical force is applied along the polarisation axis using a loudspeaker. The preload of the order of a few Newtons is required to keep the sample from vibrating out of the system. A screw thread enables both variable pre-loads and specimen sizes to be accommodated. A Kistler load cell is placed in-line with the specimen to monitor the sinusoidal load and the resulting charge is captured from the contact beneath the specimen. Charge leakage is prevented by electrical isolation of the charge collecting contact.

The system set-up for piezoelectric loss measurement is shown in Figure 2. A Solartron 1260 gain phase analyser was used as both a programmable signal generator and lock-in amplifier. A sinusoidal waveform of specified frequency and amplitude provided the drive signal for the shaker, after power amplification. The resulting sinusoidal charge and load signals from the sample were input into Kistler charge amplifiers (virtual earth and hence short circuit conditions) whose time constants were set at levels which ensured the lower cut-off frequency was below the minimum operating frequency (charge amplified by a factor of 1000 with a time constant of 10s, load unamplified with a time constant of 1s). This enabled the signals to be converted into engineering units (picocoulombs and Newtons). These signals were measured and recorded using the Solartron and associated PC software. Signals were monitored over ten cycles to provide a calculated average amplitude and phase angle and also to increase the signal to noise ratio. The sample was shielded by surrounding it with a copper sheet connected to the electrical ground to minimise electrical noise, and the entire measurement head was placed on foam pads to minimise the effects of mechanical vibrations from the surrounding environment. Solartron measurements were made in voltage “monitor”

mode, employing a feedback loop to ensure the alternating load applied to the sample was maintained at constant amplitude regardless of operating frequency or mechanical system resonances.

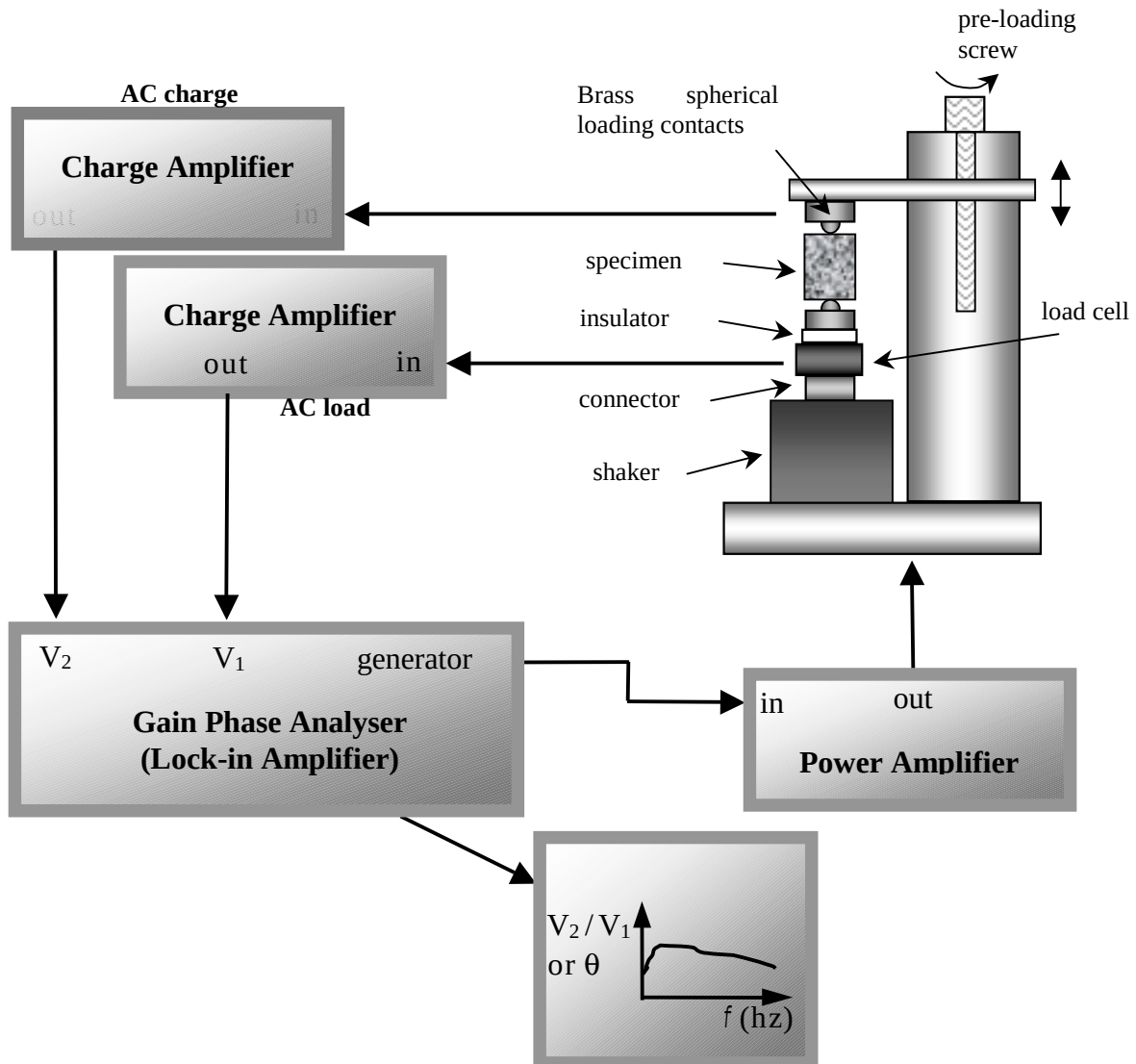


Figure 2 – Piezoelectric loss measurement system.

Investigations were carried out using 1mm thick discs of soft and hard material (PC4D and PC5H respectively) on the effects of frequency, static pre-load and AC load, in assessing the suitability of the technique to determine piezoelectric losses. Lithium niobate and quartz, low loss linear materials, were also used to distinguish the material responses from those of the measurement system.

Initially, the measurement response of the Solartron was checked over a small frequency range by using the signal generator output as both the two input signals. This ensured a known zero phase angle and voltage ratio of 1, as can effectively be seen in Figure 3.

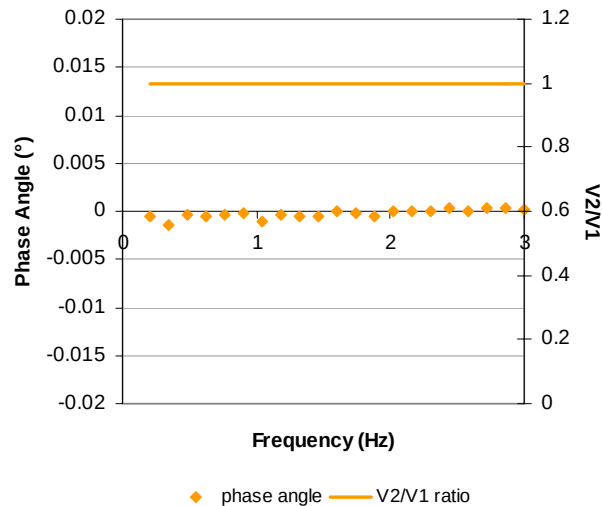


Figure 3 – Solartron system checks.

Ideally, a similar method could be used to check for any offset effects of the charge amplifiers, however, the same signal cannot be input to both. An alternative method for assessing the effect of these in the measurement circuit using nominally identical signals would need to be devised.

EFFECTS OF FREQUENCY

Frequency sweeps were carried out over the range 1-500 Hz after which mechanical resonances of the system dominate measurements. The AC load was maintained at 2N peak to peak amplitude over this frequency range.

Figure 4 (a) shows the linear-log relationship between frequency and d_{33} for both hard and soft materials. As seen in previous studies [18], a reduction in d_{33} occurs with increasing frequency for soft material, but the hard material is relatively insensitive. This is further confirmed by Figure 4 (b) which shows similar measurements carried out on low loss, linear materials (quartz and lithium niobate). These materials show a constant d_{33} over the frequency range studied. Figure 5 (a) and (b) show the phase angle losses increasing in the order: quartz < lithium niobate < PC4D < PC5H with the phase angle increasing rapidly at lower frequencies, as might be expected with increasing time for domain reorientation and motion in this region. However, as this effect is seen in all the materials over this range it may be system controlled. At higher frequencies, the dominant effects of the system resonance can be seen in both d_{33} and phase angle results, independent of material.

Some difficulties are experienced when making measurements on quartz and lithium niobate because of the low piezoelectric coefficient resulting in charge production one or two orders of magnitude lower than the non-linear materials hence a greater influence of noise, particularly for phase angle measurements.

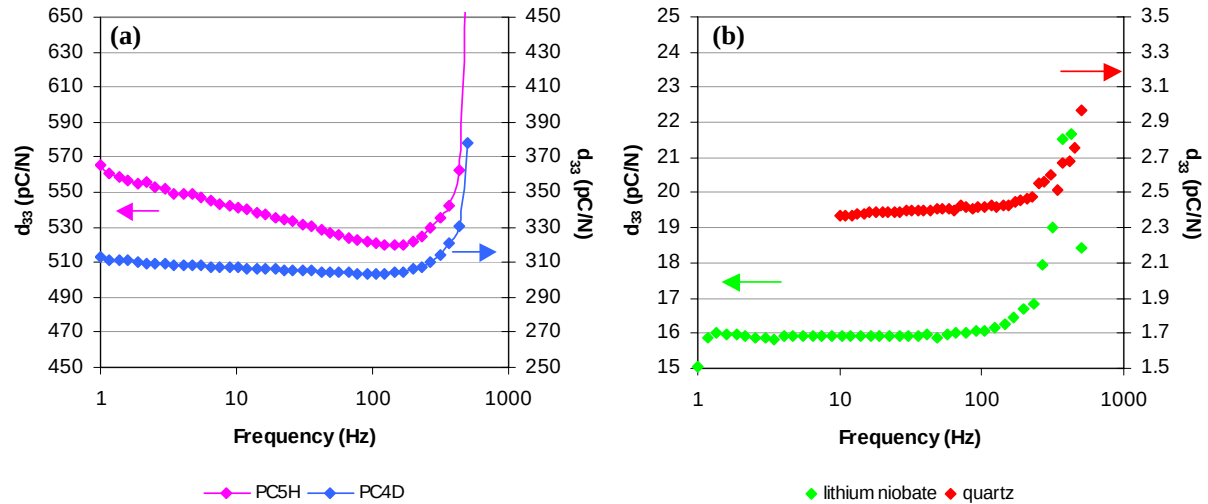


Figure 4 – Effect of frequency on piezoelectric coefficient for (a) hard and soft, and (b) linear materials.

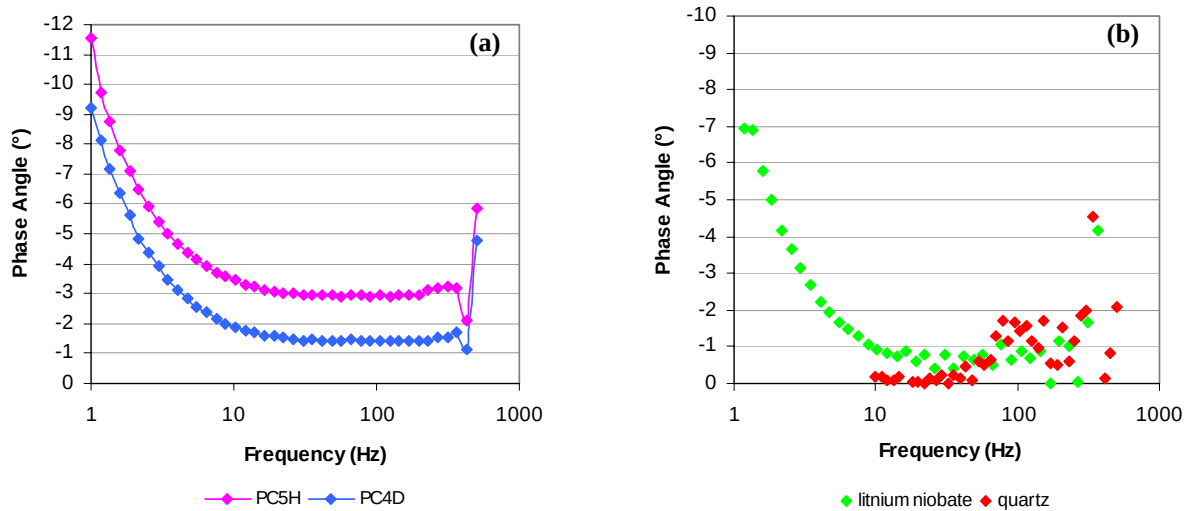


Figure 5 – Effect of frequency on phase angle losses for (a) hard and soft, and (b) linear materials.

LOW FREQUENCY REGIME

Frequency sweeps were carried out over the range 0.2-5 Hz. The AC load was maintained at 2N peak to peak amplitude over this frequency range. The charge amplifier time constants were set to ‘long’ (>1000s) for these quasi-static measurements and the results were averaged over 20 cycles. Problems were experienced with drift and noise levels which predominantly affected results at these low frequencies, as shown in Figure 6. Here, the phase angle trend contradicts that seen in Figure 5 (b) and phase angles are positive rather than negative. Further isolation of the sample from the environment (minimising charge leakage due to air currents and humidity) would help to improve these. In addition, the gain response of the power amplifier dropped sharply at very low frequency making it difficult to control the drive signal accurately, an effect which it may be possible to alleviate by using a DC amplifier for similar quasi-static tests.

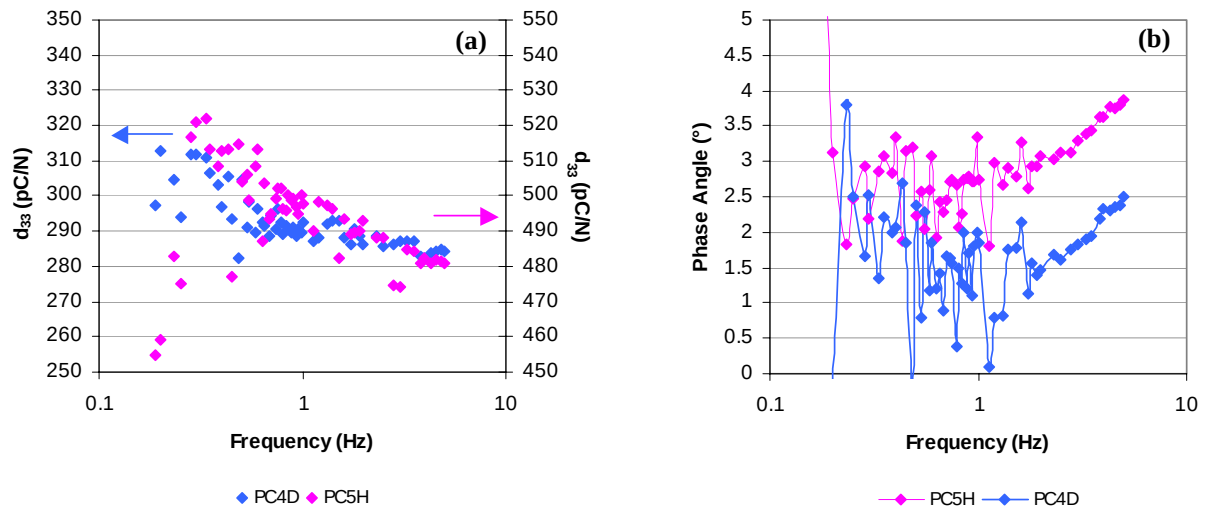


Figure 6 – Effect of frequency on: (a) piezoelectric coefficient and (b) phase angle losses for hard and soft materials.

EFFECT OF PRE-LOAD

Frequency sweeps were carried out over the range 1-500 Hz. The AC load was maintained at 2N peak to peak amplitude over this frequency range. Sufficient pre-load was applied to ensure the sample did not rattle over the frequency range and AC load amplitude studied. This was set as the base pre-load level and was increased further by turning the pre-loading screw which lowered the sample and loading configuration closer to the loudspeaker. It was also important to set the load low enough that it did not truncate the sinusoidal waveform applied by the loudspeaker. This empirical method of pre-load level application has potential for improvement by the introduction of an independent measurement of the applied pre-load e.g. strain gauge or fibre optic strain sensor. This would ensure the pre-load was accurately applied regardless of sample stiffness, thickness or surface planarity.

The results are shown in Figure 7 for a range of pre-load levels at a drive frequency of 110 Hz. The d_{33} coefficient is shown to decrease with increasing pre-load for the soft material, whereas the reverse is seen for the hard material. The effect of pre-load is also much larger for the soft material than for the hard, as seen in previous studies [18]. For the piezoelectric loss, the effect of pre load on the phase angle is larger for the soft material, as might be expected, and decreases with increasing pre-load for both hard and soft. However, the similarity between the phase plots for both materials may suggest system rather than material dependence, since in many cases the soft and hard materials exhibit different behaviour. The changes in d_{33} and phase angle due to a change in pre-load were found to be frequency independent over the range studied.

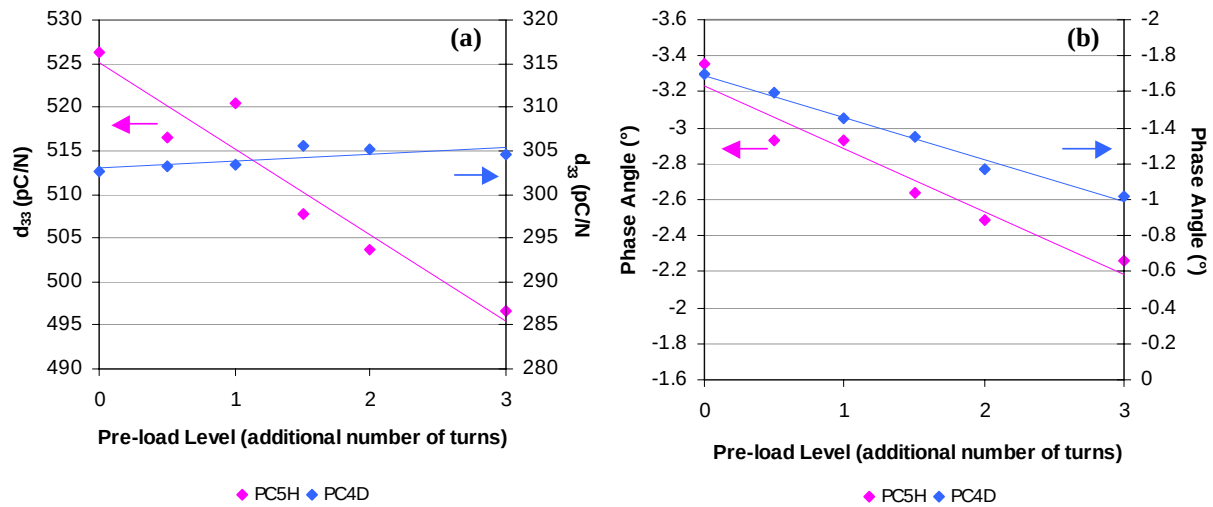


Figure 7 – Effect of static pre-load on: (a) piezoelectric coefficient and (b) phase angle losses for hard and soft materials.

EFFECT OF AC LOAD AMPLITUDE

AC load amplitude sweeps were carried out over the range 0.3-7 N peak-to-peak. The drive frequency was maintained at 110 Hz over this amplitude range, and the pre-load was sufficient to maintain the sample in the system.

Figure 8 shows the effect of increasing the applied stress amplitude on the d_{33} and the phase angle. The effect of changing AC stress amplitude is to increase the d_{33} and phase angle, and the increase is larger for the soft material than the hard. The effect is far larger than that seen with lithium niobate where both parameters are constant with amplitude (deviations at low AC amplitude are likely to be related to low charge signals and higher influence of noise due to the lower piezoelectric coefficient of this material). The phase angle loss increases with increasing stress amplitude as a greater fraction of domains become active at higher stress. Evidence of the clipping of the sinusoidal load waveform applied by the loudspeaker can be seen in the results at higher AC amplitudes, where the linear increase flattens out.

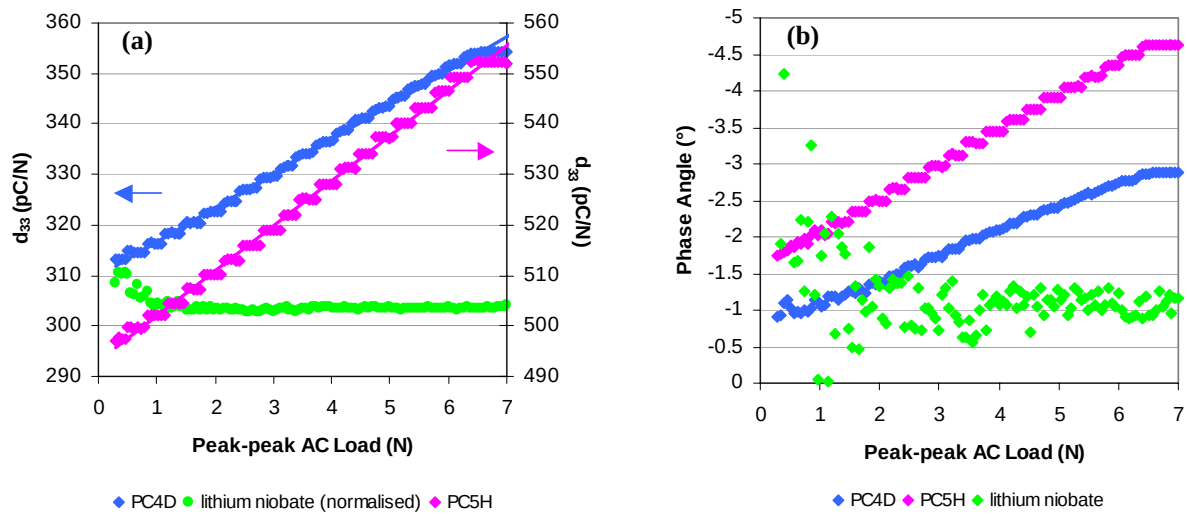


Figure 8 – Effect of AC load peak-to-peak amplitude on: (a) piezoelectric coefficient and (b) phase angle losses for hard and soft materials and lithium niobate.

INTERACTION BETWEEN AC AND DC LOADS

Experiments were carried out similar to the procedure for changing AC load amplitude but the base pre-load level was increased by turning the pre-loading screw another 1 and 2 turns.

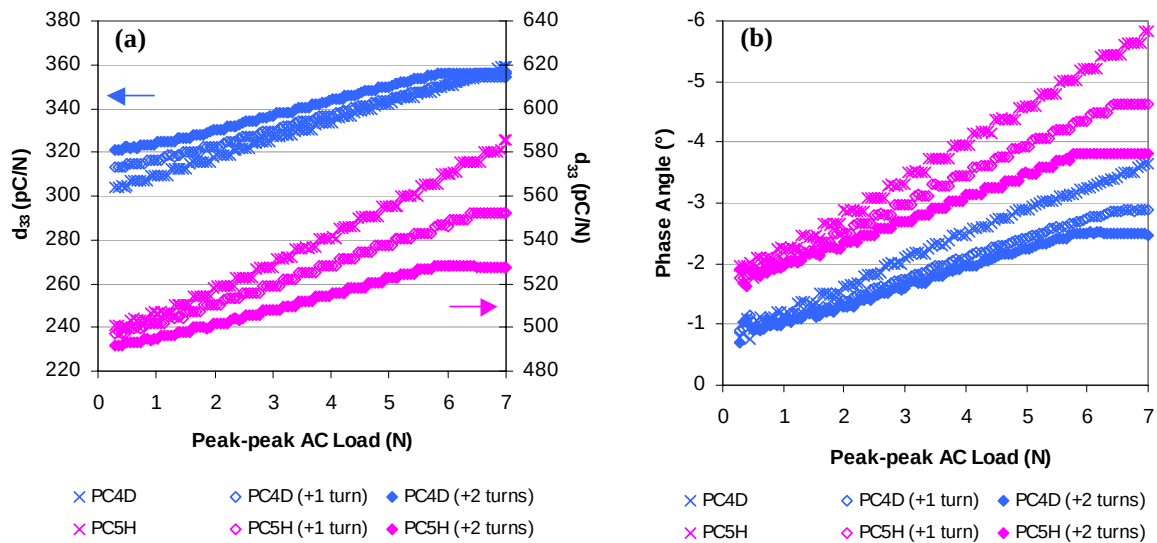


Figure 9 – Interactive effects between static pre-load and AC load peak-to-peak amplitude on: (a) piezoelectric coefficient and (b) phase angle losses for hard and soft materials.

The results from these tests are shown in Figure 9, where the effect of pre-load on d_{33} is increased for higher stress amplitudes for soft material but the opposite is true for the hard material. The phase angle is reduced as the pre-load is increased for both materials, and the effect is greater at higher stress amplitudes. Again because both hard and soft materials behave in the same manner, the system rather than the material may again predominantly influence this similarity in loss behaviour. The clipping of the shaker output at high AC amplitudes is also seen to increase with increasing pre-load.

Previous studies of piezoelectric loss using the Berlincourt method [19] have shown that the influence of mechanical resonances is more severe on the phase measurement than on the amplitude. This makes the loss measurements difficult and further refinement of the test system and instrumentation is required before the losses can be determined reliably over a wider frequency range.

Similar trends have been found for the increase in d_{33} and hysteresis or phase angle with increasing AC load amplitude, at a frequency of 1Hz and DC stress of 20MPa [14]. This also confirmed the linear change in d_{33} with the log of the drive frequency.

Measurement of Mechanical loss

As discussed, the elastic properties of piezoelectric materials are usually determined by resonance measurements, as this enables the full set of elastic constants of these anisotropic materials to be made with a few relatively simple experiments. Until relatively recently the constants determined by this method were considered real, and the out of phase components were simply ignored. Advances in the analysis of the resonance data has made it possible to account for any phase differences between stress and strain in these measurements by considering the elastic constants to have real and imaginary components. Although the resonance method is quick and simple, it suffers from two difficulties: firstly, it is difficult to do these measurements under high stresses, and secondly the measurements at resonance frequency are necessarily at high frequencies. These drawbacks are not always problems, for instance, if the aim is to predict high frequency resonator behaviour. However it is questionable whether the values gained are useful in modelling the behaviour of quasi static high load actuators, such as diesel injectors. A more conventional method of obtaining elastic property data at low frequencies and high stresses is to perform stress strain measurements, although for ceramic type samples this is generally a difficult experiment.

Initially some stress strain measurements were performed in a servo hydraulic Instron. The sample size of 6.35mm diameter meant that in order to prevent sample depolarisation or damage, loads had to be kept well below 1kN. The only available load cell for this system was 20kN and this made load control below at maximum load amplitudes of 1kN difficult.

In order to perform the mechanical measurements at lower loads and also potentially at higher frequencies, a system was constructed to apply sinusoidal loads using a piezoelectric stack actuator. A schematic of the measurement setup is shown in figure 10. In order to transfer the displacements generated by the piezo stack into a load in the sample, rather than a displacement in the system, it is necessary to eliminate any unnecessary deflections in the load train. The loading frame is a very simple, stiff arrangement, and is the stiffest part of the whole assembly. Since the piezoelectric actuator is not designed to work in tension, a DC pre-load is necessary, which is applied by the screw thread at the top of the system. The load cell sits underneath the piezo actuator, and since it is strain gauged device, is the most compliant component in the system. The overall load generating output of the system could be improved by the addition of a stiffer load measuring sensor, such as a quartz load cell, however these generally lack the DC stability of strain gauged load cells. The sample is electrically isolated from the system by two alumina plates, one which is integral to the piezo actuator, and one placed above the sample. None of the joints in the system were glued, in order to maintain high stiffness, some components in the system were held together by a screw thread, and

some simply by the mechanical DC pre load. However to aid in building up the system some hot melt glue was used around the edges to keep the stack of components stable before applying the pre load. The sample was electrically isolated from the system, but in order to make electrical contacts to perform short circuit measurements two copper shims, 150 μ m thick, were placed above and below the sample. In some initial experiments the system was used with no special attention paid to sample alignment, however it was discovered that it was possible to change the phase of the strain to the applied load by 180 degrees by simply turning the sample upside down. This meant that the strain seen by the sample strain gauge was actually a bending strain, a consequence of some non parallel surfaces. In order to accommodate misalignments a 3mm ball bearing was introduced into the load train, and this stopped the change in phase behaviour, however the measurements were still heavily dependant on the sample alignment. Because of the small sample size, it was only possible to accommodate one strain gauge. The alignment of the sample could be improved by using three strain gauges to monitor the strain and check for any bending by minimising the differences seen by the three gauges.

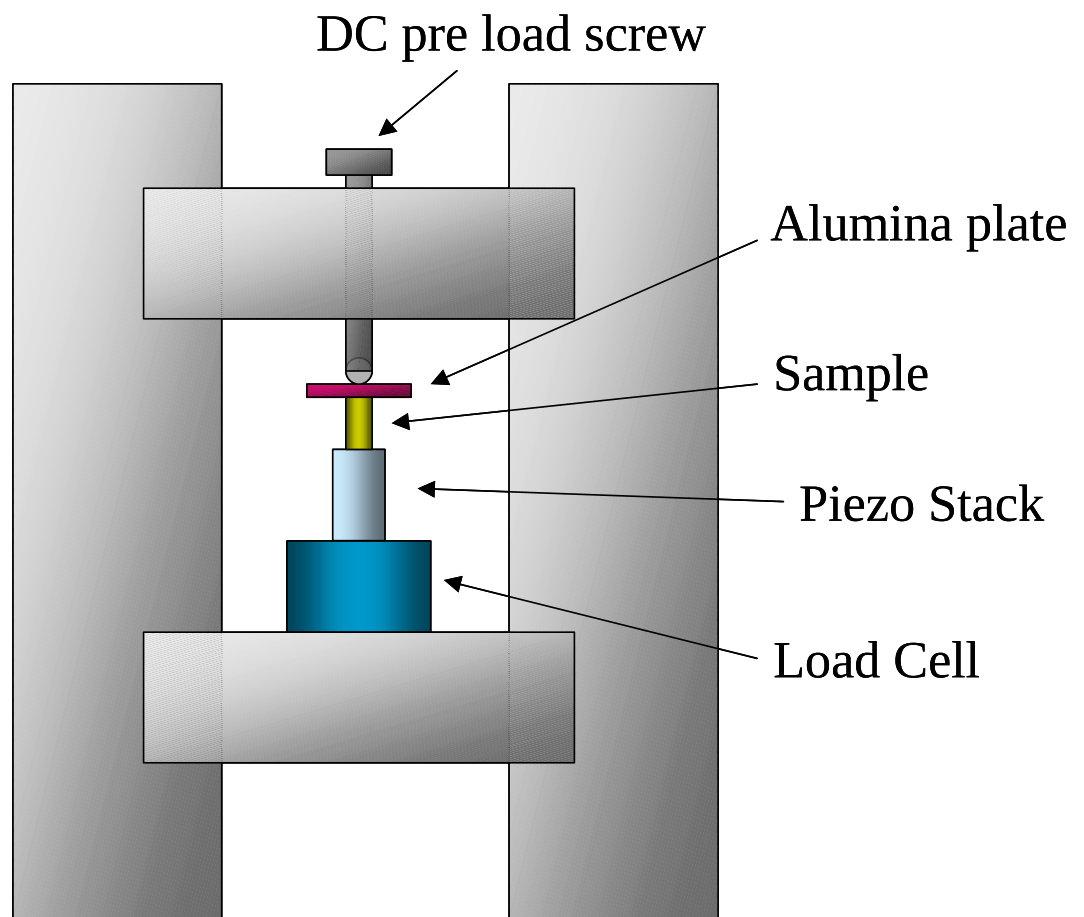


Figure 10 Schematic of the measurement setup

The piezoelectric actuator used to apply the load was a stack of multilayer actuators, totalling 30mm high and 10mm² loading area. The multilayer meant that the operating voltage of the actuator was up to 200V to produce a free displacement of around 30mm. This value was reduced when the stack was under a pre load. Although it was initially hoped that the system would be able to operate much faster than the servo hydraulic system, the current limit of the power amplifier used to drive the piezo actuator meant that at frequencies of above 200Hz the

amplifier was clipping. The Krohn-Hite 7000 amplifier has a current limit of 600mA RMS at 200V_{peak} to peak, however the piezo actuator could be driven faster given a more powerful amplifier.

The samples used in these measurements were materials that are used in piezoelectric spark generators, as these were sufficiently large to accommodate the strain gauge and also with a length to area aspect ratio to minimise friction at the sample loading interface. The materials were a hard PZT 4D and a soft PZT 5A, unfortunately the very soft 5H type compositions were not available, as they are unsuitable for spark generation.

In some preparatory experiments a lock in amplifier was used to analyse the signals, however because of problems with the signal levels generated by the strain gauge amplifier, the lock in was substituted by the Solartron 1260 gain phase analyser. This has the advantage that as well as determining the phase between two signals, in this case the load and the strain, it can also perform simple mathematical operations on the two signals. For instance, by dividing the strain by the load, it gives a number proportional to the compliance. Figure 11 shows the instrumentation set up for the measurements.

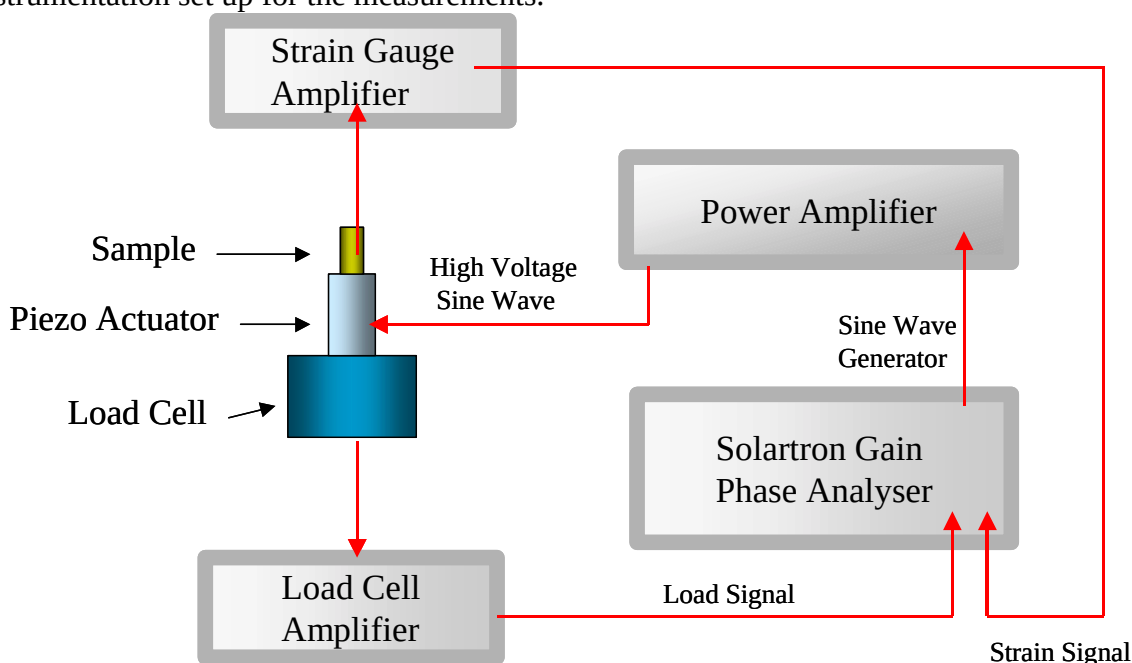


Figure 11 Instrumentation set up for mechanical compliance and loss measurements

Although the Solartron gain phase analyser can distinguish between signal at the reference frequency and noise, it is still subject to noise at the same or integer multiples of the reference frequency. This is illustrated in figure 12, which shows the compliance signal as a function of the applied load amplitude, for two frequencies, 78Hz and 100Hz. For the 100Hz signal there is a repeatable oscillation with applied load, which is because of 50Hz mains noise interfering with the reference frequency. This problem can be eliminated either by improving the shielding of the system, or by working away from the mains frequency. As can be seen, working at 78Hz greatly improves the signal, and is a simpler method of removing the noise.

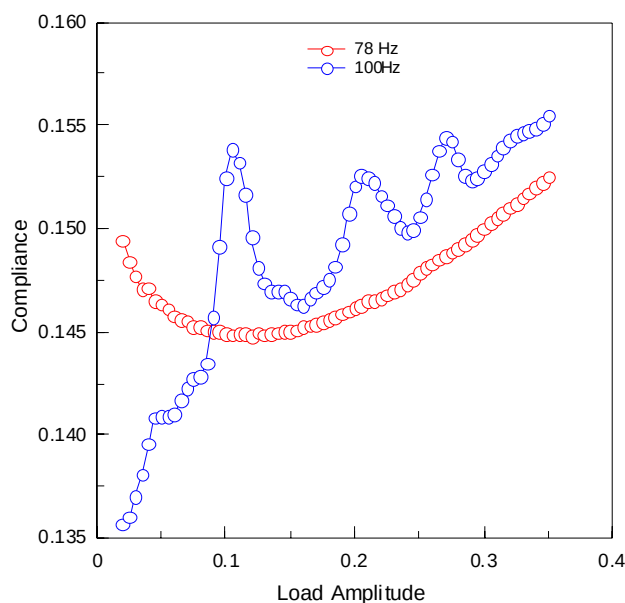


Figure 12 Compliance signal as a function of the applied load amplitude, for two frequencies, 78Hz and 100Hz.

It was clear from some of the early investigations that the absolute values for the compliance and loss results depended greatly on the sample alignment, and so direct comparisons between measurements on two separate samples would be difficult. However by changing some parameters during the experiment, without removing the sample, such as applied load amplitude, frequency, or open/short circuit conditions some measure of how samples behave differently could be obtained. Figure 13 shows the amplitude sweeps for the hard and soft material, under open and short circuit conditions. From this it can be seen that going from short to open circuit conditions the compliance is reduced, i.e the material gets stiffer in the open circuit, as the charge build up creates an opposing force to deformation. The compliance is reduced by a factor of around 1.6, which is similar to the factor of 2 times seen for the resonance experiments. The shape of the compliance versus applied load curves for the two materials are different, with the hard reaches a maximum roughly in the centre of the sweep, whereas the soft reaches a maximum at the end of the sweep. The loss measurements also show significant differences between open and short circuit conditions, going from negative to positive. This observation is obviously tempered by the fact that there are no absolute values in these experiments, and that the zero loss point has not been defined. However it does show that the materials behave measurably different, and that an improved experiment would yield interesting information.

The effect of frequency on the measured compliance and loss also shows similar effects between the hard and soft materials and the open and short circuit conditions. The results show a small deviation at around 50Hz because of the mains noise. In an attempt to ascertain the zero loss conditions an alumina sample was tested, and the frequency response of the loss was similar to the loss for the short circuit measurements, i.e. very linear with frequency. Again this indicates that the different loss results seen in the open circuit conditions are material related and deserve further investigation.

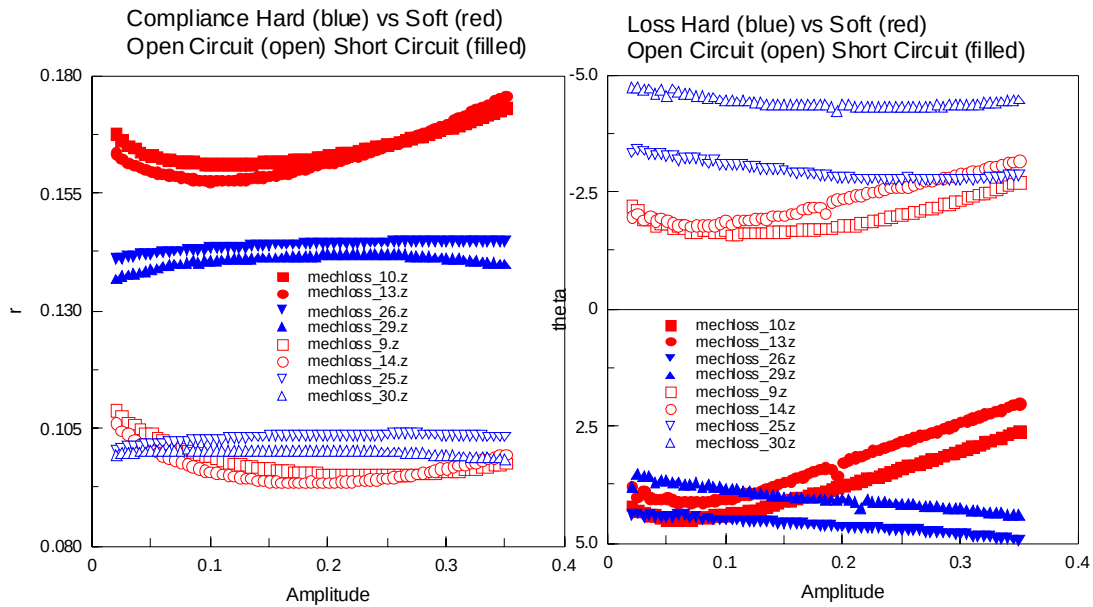


Figure 13 Amplitude sweeps for the hard and soft material, under open and short circuit conditions.

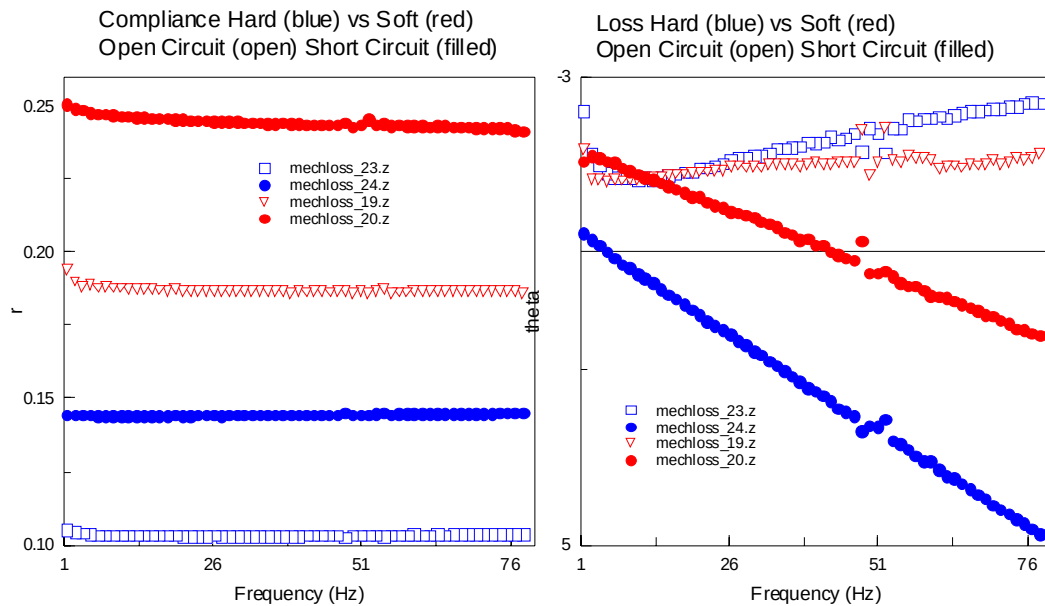


Figure 14 Frequency sweeps for the hard and soft material, under open and short circuit conditions.

There are few published results that describe the effect of AC mechanical loading amplitude on the mechanical compliance and loss of piezoelectric materials. Most workers performing stress strain experiments tend to concentrate on the high stress range where depoling occurs [20,21]. Fett and Munz, have critically compared Young's modulus measurements using stress-strain and resonance and shown them to agree well [22]. They assume that the value is

a constant, and do not consider the effect of strain amplitude. R Gerson [23] has looked at Young's modulus and mechanical Q of a resonant transducer as a function of increasing electrical drive level, i.e increasing strain amplitude, and shown that the modulus and Q both decrease, the amount depending on the material. Gerthsen et al. [24] have shown using resonance experiments that the mechanical loss increases with increased electrical loading and is linearly related to the dielectric loss. Recently Priya et al [25], also using electrically induced resonance, have shown that the compliance increases linearly with the square of the strain amplitude, and that the mechanical Q decreases. Uchino and Hirose [3] have measured the mechanical loss under short circuit conditions, using similar stress strain measurements to this work, and noted that the elastic loss is rather insensitive to the applied compressive stress.

Conclusion

A novel system for measuring the phase angle between applied stress and generated charge has been developed based on a Berlincourt test. The results for the piezoelectric coefficient d_{33} agree well with previous findings, the coefficient increases with increasing applied AC stress amplitude, and the behaviour for hard and soft materials differs with respect to DC preload and changes in frequency. The piezoelectric loss results show an increase in loss with increasing AC amplitude for hard and soft, however the increase in loss seen at low frequencies is seen in all materials including Lithium Niobate, suggest that this is an experimental artefact. The absolute values of the loss are hard to quantify because of pinning down the losses associated with the measurement system, however the relative values of the loss are as expected, soft > hard > lithium niobate > quartz.

The elastic modulus measurements were more difficult to quantify because of bending of the sample, giving rise to arbitrary initial phase angles that could not be corrected. However, by varying the sample environment, such as open and short circuit conditions, it was possible to show interesting material dependant effects. As expected the elastic modulus increases by a factor of around 2 on going from short circuit to open circuit conditions, and the loss also shows large changes on changing the boundary conditions. This test could be improved by adding more strain gauges to aid in sample alignment, however the fundamental flaw in the strain gauge method is that it only measures the strain at the surface. It should also possible to increase the reliability of the results by using a miniature bend test, and measure actual bending displacements, rather than surface strain through a strain gauge.

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Glossary

- ω = Angular Frequency (Radians per second)
 $\tan \delta$ = Dielectric Dissipation Factor
 ϵ_r^* = Complex Relative Permittivity
 ϵ_r' = Real part of complex relative permittivity
 ϵ_r'' = Imaginary part of complex relative permittivity
 ϵ_{ij} = Absolute Permittivity
 ϵ_0 = Permittivity of free space (Farads per metre)
 C = Capacitance (Farad)
 V = Voltage (Volts)
 E = Electric Field Strength
 D = Dielectric Displacement
 P = Polarisation
 X = Stress
 x = Strain
 d_{ij} = Piezoelectric Charge (Strain) Constant (C/N) or (m/V)
 s_{ij} = Elastic Compliance Constant (metres-squared per Newton)
 h_{ij} = Piezoelectric constant, electric field/strain at constant charge or stress/charge density at constant strain (V/m)
 ϕ^* = The phase delay between stress and strain under constant electric field (short circuit)
 δ^* = The phase delay between electric displacement and electric field under a constant stress X (zero stress, usually measured at 1kHz)
 θ^* = The phase delay between strain and electric field at zero stress or the phase delay between charge and stress at constant electric field (short circuit)
 ϕ = The phase delay between stress and strain under constant electric displacement D (open circuit)
 δ = The phase delay between electric displacement and electric field under constant strain x (zero strain, completely clamped, usually measured far above resonance)
 θ = The phase delay between open circuit field and applied stress or the phase delay between applied charge and strain developed
 Q_m = Mechanical Quality Factor