

# REPORT

## **Measurement methods for investigating degradation in piezoelectric ceramics**

Mark Stewart

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# MEASUREMENT METHODS FOR INVESTIGATING DEGRADATION IN PIEZOELECTRIC CERAMICS

M. Stewart  
Centre for Materials Measurement and Technology  
National Physical Laboratory  
Teddington  
Middlesex  
United Kingdom  
TW11 0LW

## ABSTRACT

Methods for testing the fatigue induced degradation of piezoelectric ceramics have been investigated. Measurements under cyclic electrical loading have proven difficult due to dielectric losses leading to sample heating, but several methods to overcome this have been identified. Cyclic mechanical loading methods at rates in excess of 100Hz have also been investigated, using a resonant electromagnetic loading system and a conventional servo-hydraulic system. Examination of the mechanically fatigued samples has shown a reduction in the electromechanical coupling coefficient,  $k_{\text{eff}}$ , and the stress relief mechanism is brought about by 90° domain switching.

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National Physical Laboratory  
Teddington, Middlesex, UK, TW11 0LW

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## 1 INTRODUCTION

There are several mechanisms by which the performance of piezoelectric ceramics can degrade and thus limit the use of these materials. The ultimate static performance is limited by the dielectric breakdown and its mechanical counterpart the mechanical strength. However failure and loss in performance can occur at loads well below the static failure due to a combination of time and cyclic loading. Ageing is the loss of performance with time in the absence of any mechanical or electrical loads and is simply quantified by measuring the various piezoelectric parameters at intervals after poling and is described as a percentage loss with time. Time dependant dielectric breakdown is a phenomena whereby dielectric breakdown occurs at decreased loads due to the long term application of a DC load<sup>1</sup>. Most of these quasi static phenomena are relatively well understood<sup>12,3,4</sup> and models for the behaviour have been developed, however as the applications for these ceramics have pushed up the operating loads, the degradation due to cyclic loading has become more important. Therefore new test methods need to be developed to simulate these conditions so that the degradation can be measured.

The industrial use of these materials are many and varied and each application is unique with its own unique operating conditions and environment. This diversity of applications would be difficult to cover comprehensively, however they do fall neatly into two categories as either sensors or actuators. In general an actuator will see an applied voltage operating in order to give a displacement against some sort of static load, whilst a sensor will see an applied mechanical load in order to generate a charge or voltage. These categories are not rigid, for example a vibration damping actuator will also see an alternating mechanical load in addition to the electrical driving voltage. However the experimental simulation of such an application would be complex and it might be easier to extrapolate from separate electrical and simple mechanical loading experiments.

## 2 ELECTRICAL LOADING (ACTUATORS)

As has been previously stated the static behaviour of piezoelectric ceramics is reasonably well understood and the area of most concern is fatigue after repeated electrical cycling. Although most actuators will be required to perform some work in moving a load the simplest form of experiment to carry out is the unloaded case. Here the sample is placed under an AC load for an increasing number of cycles, whilst measuring the performance of the ceramic, either continuously or at predetermined intervals. There is a choice of whether to measure the displacement, or the input current at a fixed voltage, but for an actuator the displacement output seems the most appropriate measure. The experimental set-up is shown in figure 1, where the sample, in the form of a disk, sits held on a brass vacuum holder by atmospheric pressure. The brass holder is also the high voltage electrode. An AC voltage is applied across the thickness of the disk and the displacement is constantly measured by a fibre optic displacement probe. In an attempt to produce a fatigue in a reasonable time a 6kV field @ 200Hz was applied across the 2mm thick PC4 sample.

Initial results indicated that the displacement output decreased rapidly with increasing number of cycles, however closer investigation showed this was not due to fatigue. Figure 2 shows the output of the fibre optic probe after applying the voltage for 10 minutes and then turning the load off, but continuing to measure the probe output. The form of the curve is in fact caused by the sample and the holder expanding due to dielectric sample heating and then contracting

on cooling, and this is confirmed by the thermal image of a similar experiment after 2 minutes of applied load (figure 3). The 'hot' spots in the image are the sample, the PTFE holder and the curled ground lead. Separate measurements with a thermocouple shows the sample temperature increase is only around 10°C, but this is enough to cause significant thermal expansion.

The design of the sample jig which was built for high speed displacement field hysteresis measurements could be improved by replacement of the PTFE with a lower expansion material, however there are several other points to consider with respect to better experimental design. Firstly thermal expansion, as the sample self heats, causes the distance between sample and fibre optic probe to decrease in such a way that the output from the fibre optic probe ceases to be linear. This could be overcome by using laser interferometric measurements, for increased range and resolution, or by performing measurements when the sample has cooled after a no load period.

Another improvement would be to monitor the current input for a fixed applied voltage, although this could also be affected by the change in sample temperature.

The sample heating is not easy to quantify since it depends on many material independent factors such as the sample size and the thermal load of the sample surroundings, and also many of the piezoelectric properties are sensitive to changes in temperature of this magnitude. One way of reducing the problem would be to use only DC half cycles<sup>5</sup>, but this does not give the switching in field which could be the cause of the fatigue. Another option used recently<sup>6</sup> would be to pulse the AC field in short bursts in order to prevent the self heating problem. The applied voltage amplitude and frequency could also be reduced in order to reduce the heating, but again this might be not enough to produce the fatigue behaviour, and the application of a mechanical load should reduce the field needed to produce degradation behaviour.

In summary this problem has highlighted the need to monitor if not control the sample temperature during future electrical fatigue experiments.

Some preliminary experiments of electrical loading under applied mechanical loads were also performed. In work on displacement measurements under different applied loads it was found that the simplest method of applying a mechanical load to a sample was by using a spring system rather than just a dead load. The spring constant of the loading system is arranged such that the small displacements caused by the piezoelectric strain do not cause a significant change in the applied load.

In an attempt to mirror some of the behaviour seen in the mechanical loading experiments, samples were clamped in a stiff testing system and an AC electrical load applied. This generates a cyclic stress in the system and the amplitude of the load, and also the applied voltage and current were monitored throughout the experiment. Figure 4 shows the load generated decreasing with time, and the form of this degradation is similar to that seen in the mechanical loading experiments (discussed later). The temperature of the sample was not monitored during the experiment and some of the behaviour, particularly the change in resistance of the sample, could be attributed to thermal effects. This again highlights the need for careful experimental techniques in order to separate the thermal and fatigue effects in these experiments.

### 3 MECHANICAL LOADING (SENSORS)

As is the case for electrical loading, the concern for mechanical loading is the degradation in performance after repeated numbers of loading cycles. So that experiments could be conducted in a suitable length of time, various methods of applying the alternating mechanical loads at high frequency were investigated.

Conventional ballscrew-driven materials testing machines only operate in the range of a few Hertz, although servo-hydraulic actuators can push the range up to around 100Hz. For both systems large loads can be easily applied. Other methods investigated were an electric motor driving a cam, where the cam translated the rotary motion into a linear one whilst also amplifying the frequency six fold. A prototype system was capable of producing a sinusoidal load of 200N at about 200Hz, but with more substantial hardware the loads could have been driven much higher, however wear of the system might have caused problems. A pneumatic loading system was also investigated, in the form of an air hammer driving into a spring damping system to modify the impact behaviour of the hammer. This system was only capable of delivering 50N at around 60Hz, and scaling up of the system was not thought to be feasible. Both these systems suffer from the same problem in that they are generally producing medium loads over large displacements (in the order of mms), whereas for the piezoelectric testing system the large loads are needed only over a few microns. The mechanical translation from large displacement-low load to large load-small displacement is often inefficient and leads to losses through wear, noise and heat.

Obviously it would be better to use a loading system which produced displacements over the range of interest, and obvious candidates for this would be piezoelectric or magnetostrictive actuators. Indeed in a recent study<sup>7</sup> piezoelectric actuators have been used to generate stresses in piezoelectric ceramics, and in principle if the loads generated are monitored there should be no problems. However it was felt that some other methods should be used first to understand the behaviour before using them in a measurement system. The most promising system for applying the loads was using an electromagnetic actuator. Small load high frequency loud speaker type actuators are often used in vibration studies and are also be used in Berlincourt type  $d_{33}$  meters, however the system finally used was an Amsler Vibraphore which uses an electromagnet to generate loads of up to 10kN at up to 300Hz.

The system is shown schematically in figure 5, and the high loads and frequency are achieved by operating the system at resonance. This is achieved by firstly generating an impulse within the system by means of a DC coil, and then resonance is maintained by an AC coil which uses the load cell feedback to lock on to the frequency. This has drawbacks in that it is difficult to start up, sometimes it takes several minutes before it is stable, and also there is little choice over the frequency of operation. The sample sits between two hardened steel anvils and is isolated from the system by two alumina spacers, so that experiments can be carried out in either short circuit (charge density mode) or open circuit conditions (voltage mode).

For all the tests the amplitude of the mechanical input and also the amplitude of the piezoelectric output was monitored continuously and the ratio of these was plotted against the number of cycles (figure 6). The mechanical loads were such that the sample was always under compression, and so the sinusoidal loads needed an offset to maintain the compression. Tests

were carried out at amplitudes up to 20kN on a 10mm diameter sample, with a mean load of 10kN, and at a frequency of 230Hz tests were conducted to over  $10^8$  cycles. The results all showed a similar logarithmic drop off in piezoelectric output with increasing number of cycles, and at the higher loads led to mechanical failure of the sample, often reducing the sample to powder.

After investigation into the cause of the noise on the output traces it was discovered that by inserting a universal joint in the loading train to help align non parallel sided samples, the previously observed fracture behaviour could not be reproduced, and that it was caused by sample misalignment and uneven loading.

The nature of the degradation was such that there is a rapid drop in piezoelectric output over the first few thousand cycles, and this is the time during which the Amsler Vibraphore is unstable whilst resonance is achieved. In order to investigate the behaviour in the early stages a conventional servo-hydraulic Instron testing machine was used, which although has a reduced frequency range, can be controlled more precisely. Figure 7 shows a the degradation in performance is similar to the Amsler experiments, but the plot of the load input over this period shows that the load input has also changed. There is a feedback system on the Instron to maintain a constant load, but this can be affected by the stiffness of the sample, and the sample holder. This result indicates that the sample stiffness is changing with increased loading cycles. This could be due to the bedding down of components such as the electrodes, indeed the electrode surfaces at the end of the experiment have a polished appearance, or it could be due to the change in elastic stiffness of the piezoceramic.

Further investigation into the mechanism of this degradation show that this occurs in both poled and thermally depoled samples, (Figure 8), although the magnitude of the depoled samples' output is greatly reduced. The form of degradation is also similar for open circuit and short circuit conditions.

Examination of the samples after mechanical cycling also show that permanent changes have taken place in the samples. The electromechanical coupling coefficient  $k_{\text{eff}}$  of some of the mechanically cycled samples was calculated from measurements of the radial resonance of the disks, and are given in table 1 along with their mechanical history. It can be seen that  $k_{\text{eff}}$  is reduced with increasing loads and number of cycles, an indication that the material is gradually depolarising.

Microstructural investigation of the samples did not reveal any evidence of damage, such as microcracking, however X-ray diffraction showed significant differences between as received and mechanically cycled samples. Figure 9 shows the relative intensities of the 002 and 200 peaks of the tetragonal PZT phase for a poled sample, a thermally depoled, and a heavily fatigued sample, 10kN for  $8.5 \times 10^7$  cycles. It can be seen that the ratio of the two peaks for the cycled sample are similar to the thermally depoled sample, and many of the 002 oriented grains have switched to 200 orientation. This  $90^\circ$  switching should be accompanied by a change in length since the 'a' direction is shorter than the 'c' direction, and this would seem favourable to relieve the large compressive stresses placed upon the sample.

The effect of sample heating due to losses in the sample was highlighted for the electrical loading experiments, however the temperature change due to mechanical loading has not yet been investigated. In similar experiments on magnetostrictive materials using the same sort of loading conditions the sample was seen to rise by about  $10^\circ\text{C}$  in the first few minutes and then stabilise as the thermal equilibrium was reached. Future work will have to examine if this is the case for mechanically loading piezoceramics.

#### 4 CONCLUSIONS

These preliminary experiments have demonstrated the experimental techniques and methods needed to perform high stress fatigue measurements on piezoceramics. For the case of electrical loading it is clear that close attention must be paid to self heating effects in order to deconvolute them from the effects of fatigue. When an experiment is being continuously monitored the change in properties due to sample temperature change must be considered by reference to the room temperature property, and the sample must be examined to see if it has undergone a permanent change.

In the mechanical loading experiments various methods for applying high stresses at high frequency on piezoelectric ceramics, with a resonating electromagnetic system and servo hydraulic test system being the most useful. Initial experiments where the samples failed catastrophically have shown the need to consider sample alignment, and this was compensated for by the inclusion of a universal joint. Investigations into the fatigue in a hard PZT, PC4, by X-ray diffraction have shown that depoling of the material occurs by 90° domain switching which tries to relieve the compressive stresses produced by mechanical loading. The depoling increases with time, as measured by the shift in resonance, however more work is needed to see if this is consistent with the change in charge or voltage output measured during the cycling, and how this varies with other piezoceramics.

#### 5 ACKNOWLEDGEMENTS

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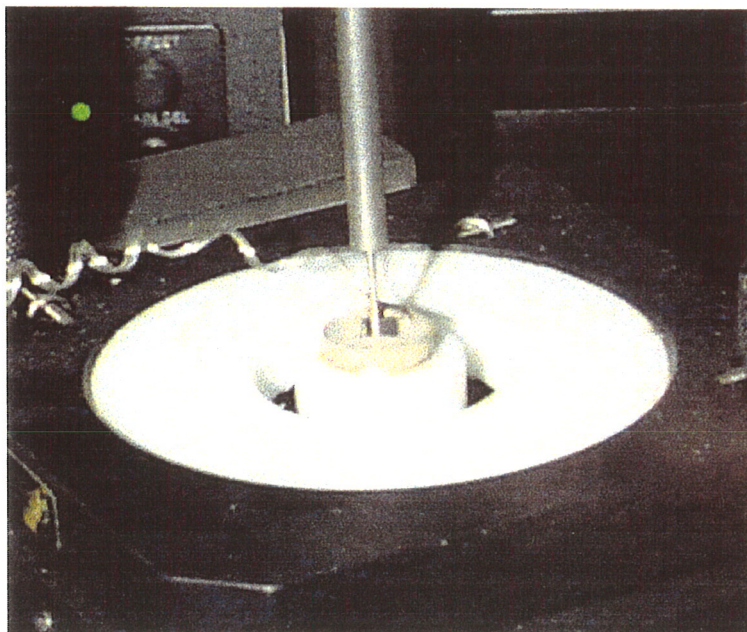


Figure 1. Fibre optic displacement measurement set-up. The PZT disk 10mm diameter 2mm thick sits on a PTFE shielded brass electrode. The sample has a small piece of reflective tape attached to increase reflectance, and the ground lead is soldered onto the sample.

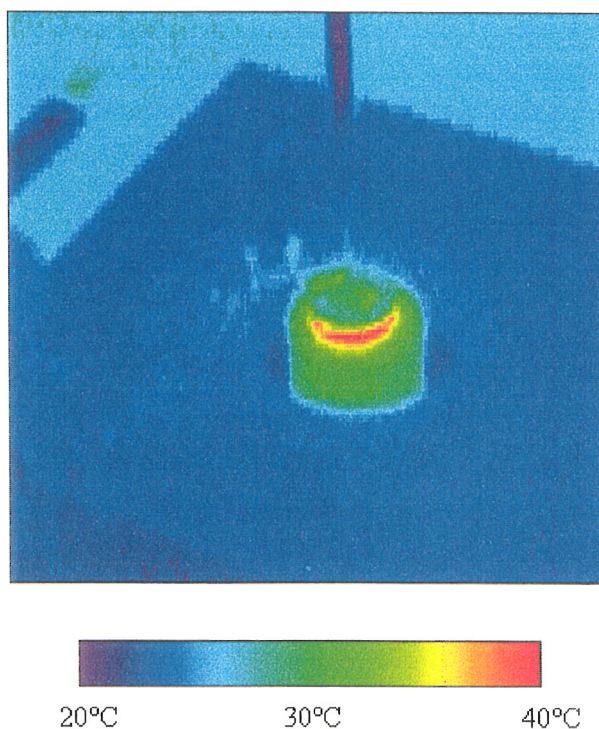


Figure 3. Thermal image of experimental set-up in figure 1, after applying a 6kV p-p AC field at 200Hz for 2 minutes. The sample itself is the hottest region, but the PTFE sample holder has also increased in temperature.



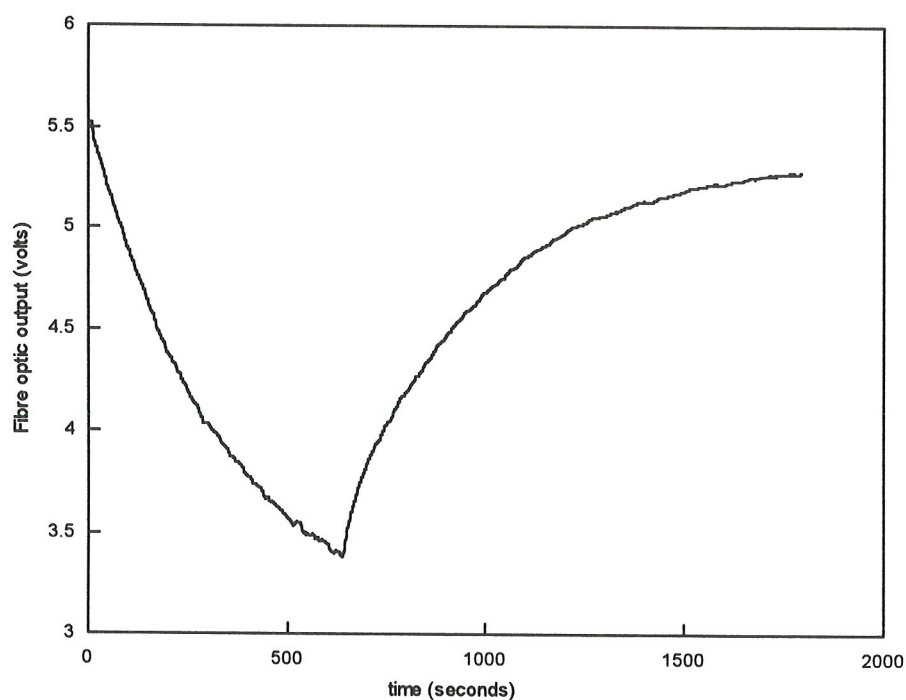


Figure 2. Fibre optic probe output for sample set-up shown in figure 1. An AC voltage of 6kV p-p at 200Hz was applied to a PC4 disk, 10mm diameter, 2mm thick, for 2 minutes and then turned off. The linear region for these probes is between 4 and 6 volts, and the AC component of the displacement is of the order of a few hundred millivolts on top of this DC signal.

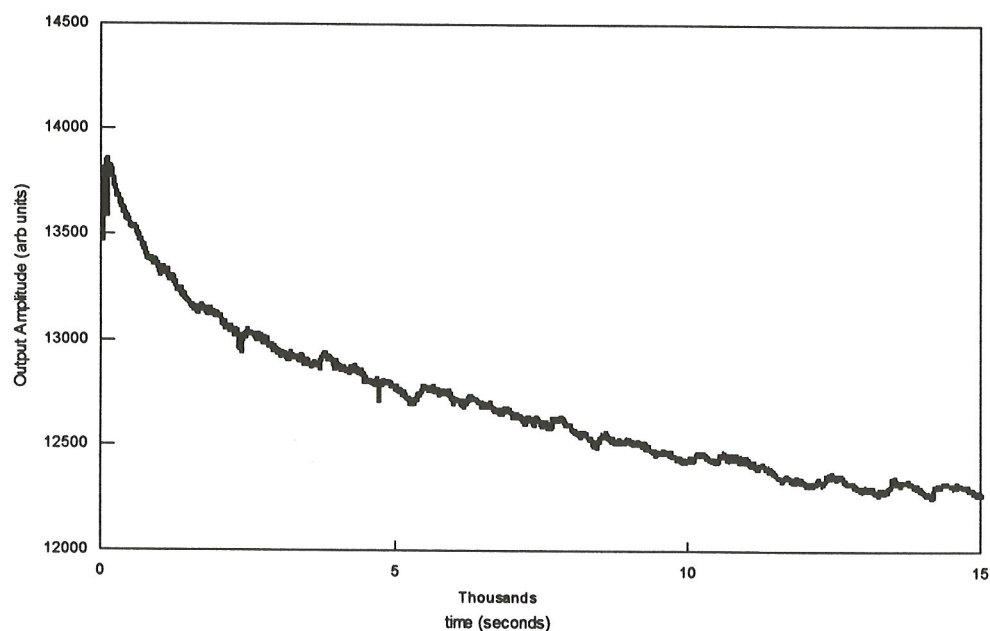


Figure 4. PC4 sample, 10mm diameter, 2mm thick, placed in stiff loading system, with 7kN load applied. A 6kV AC electrical load applied at 260Hz, and the load output of the system is continuously monitored.

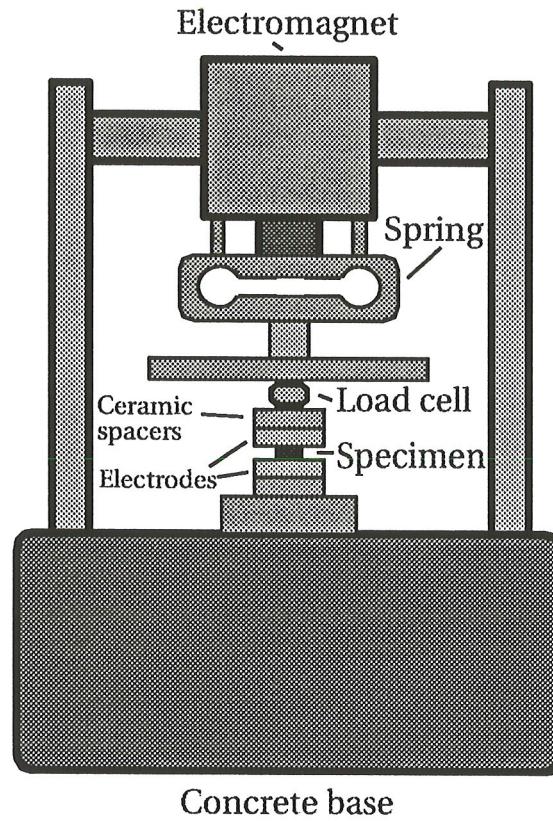


Figure 5. Schematic of Amsler Vibraphore 10-ton resonant loading system.

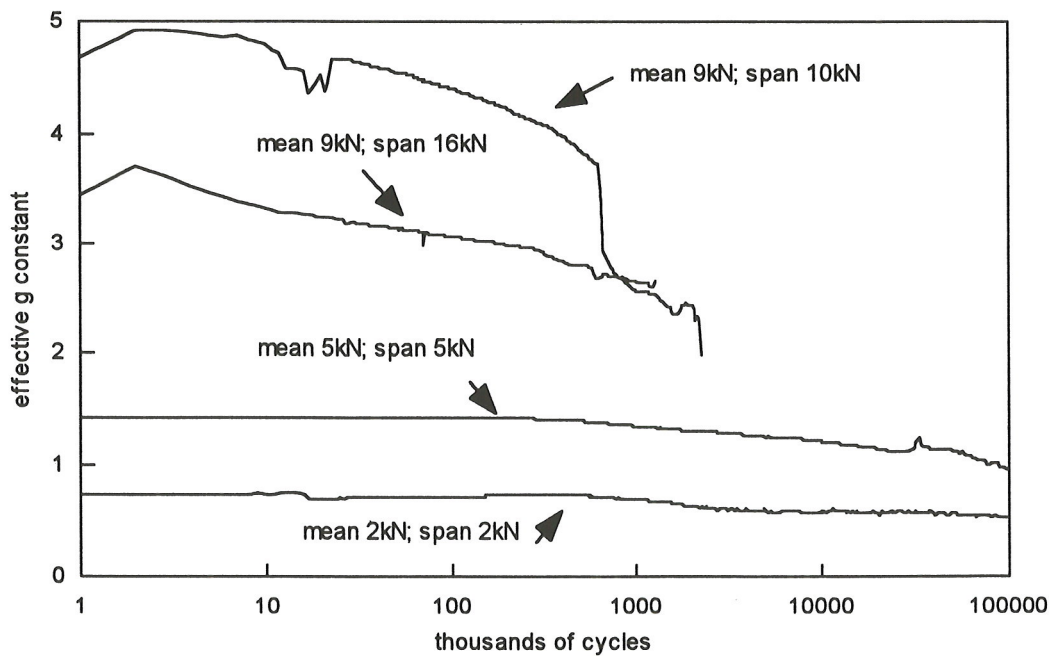


Figure 6. Fatigue loading in Amsler vibraphore system of PC4 disks. The effective g constant is calculated by dividing open circuit voltage output of the sample by the voltage output of the load cell.

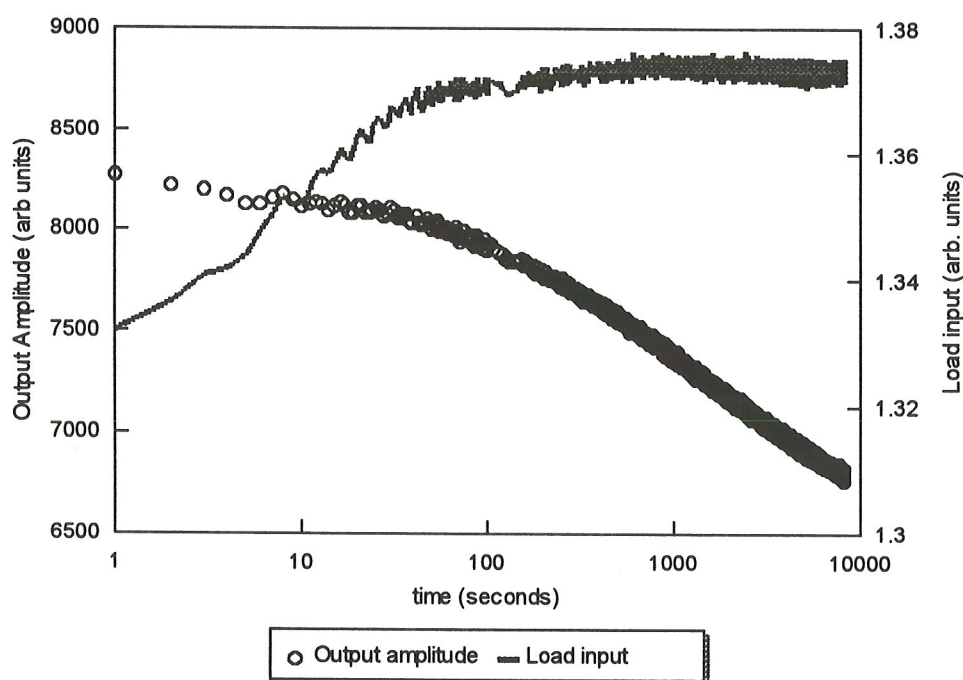


Figure 7. Fatigue experiment in conventional servo-hydraulic testing machine. Sample PC4 disk 10mm diameter, 1mm thick, mean load 10kN, span 10kN at 100Hz.

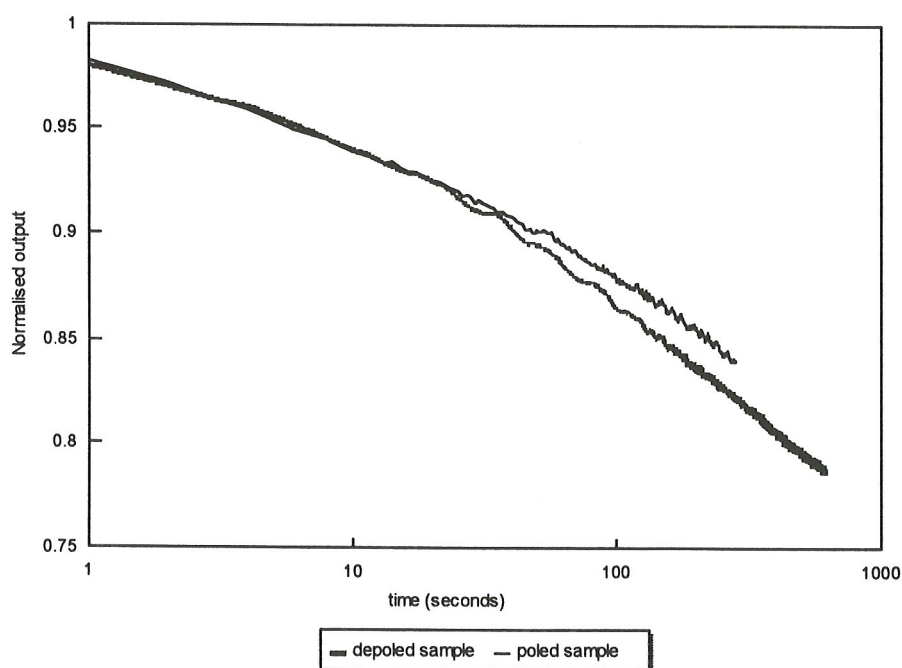


Figure 8. Fatigue of poled and thermally depoled PC4 disk samples 10mm diameter, 1mm thick, 20kN mean load, 10kN amplitude at 100Hz. The open circuit output of the samples has been normalised, the output of the poled sample is several orders of magnitude greater than the thermally depoled sample.



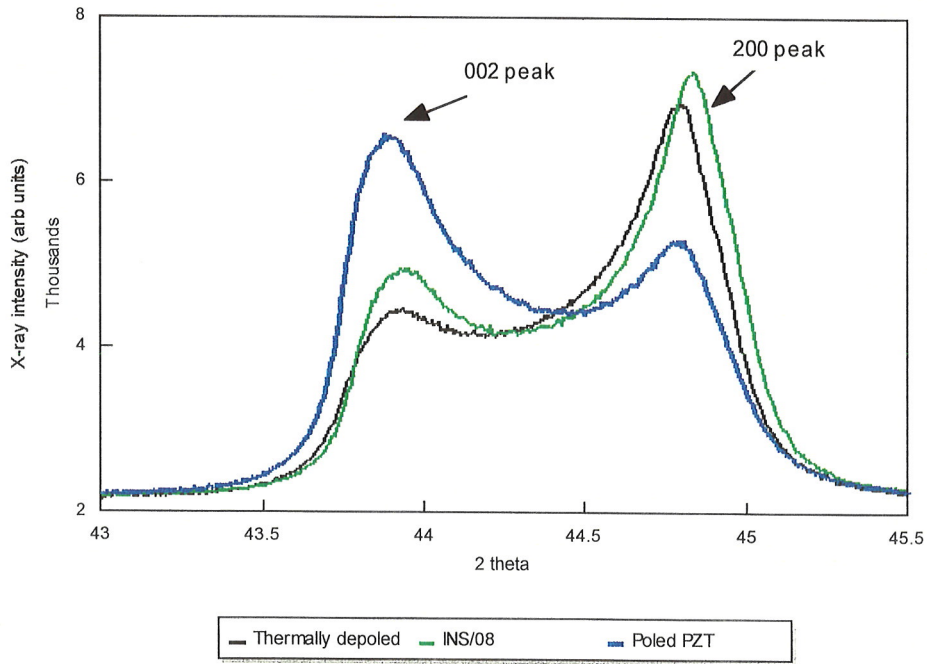


Figure 9. X-ray diffraction pattern of three PC4 samples, in the as-received state, thermally depoled, and after mechanical fatigue for  $8.5 \times 10^7$  cycles at 10kN. The PZT is tetragonal and the first peak is the [002] c-axis, and the second peak the [200] a-axis.

| sample | $k_{\text{eff}}$ | Load | No. of cycles |
|--------|------------------|------|---------------|
| klq    | 0.52             | none | as received   |
| ins03  | 0.45             | 10kn | 4E+05         |
| ins02  | 0.49             | 10kn | 2.1E+05       |
| ins01  | 0.48             | 10kn | 2E+05         |
| ins07  | 0.36             | 20kn | 4E+05         |
| ins08  | 0.42             | 10kn | 8.57E+07      |

Table 1. Effect of mechanical cycling on radial resonance of PC4 samples,  $k_{\text{eff}}$  is the mechanical coupling coefficient.

