

Real Time Evaluation of Piezoelectric Materials

Dr Markys G Cain, Dr Mark Stewart & Dr Mark Gee

January 1999

REAL TIME EVALUATION OF PIEZOELECTRIC MATERIALS

M G Cain, M Stewart, & M G Gee
Centre for Materials Measurement and Technology
National Physical Laboratory
Queens Road, Teddington
Middlesex, TW11 0LW UK

Summary

As part of the DTI programme on the development of test methods for characterisation of advanced materials, project CAM 7 *Electroactive materials properties under conditions of high stress or stress rate*, has the overall aim of defining and improving the measuring framework for electroactive materials which will enable them to be used with greater confidence by UK industry.

This report describes an investigation of the feasibility of developing robust real-time evaluation techniques for the assessment of fatigue of electroceramic materials:

1. The first technique of acoustic emission generation and detection within piezoelectric materials has been successfully demonstrated as a means of assessing internal physical processes occurring in multilayer materials and monolithic materials when driven at high electrical fields.
2. The second method of directly observing microcracks within materials or devices has been investigated. Optical imaging techniques are potentially capable of detecting micro-cracks, greater than 20-30 μm in piezoelectric ceramics.
3. The third technique of thermal imaging to monitor degradation in piezoelectric multilayer devices was not definitive, due to the constraints imposed upon this measurement method by the camera system that was used.

The merits of each technique are described and representative data compared for each method.

NPL Report CMMT(A) 149

© Crown Copyright, 1999
Reproduced by permission of the Controller of HMSO

ISSN 1361-4061

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

No extracts from this report may be reproduced without the prior written permission of
the Managing Director, National Physical Laboratory
The source must be acknowledged.

Approved on behalf of the Managing Director, NPL, by
Dr C Lea, Head of Centre, Centre for Materials Measurement and Technology

Table of Contents

<u>AIMS</u>	2
OBJECTIVES	2
<u>EXECUTIVE SUMMARY</u>	3
TECHNIQUES	3
<u>MONITORING DEGRADATION IN PIEZOELECTRIC MATERIALS</u>	5
<u>TECHNIQUE 1: ACOUSTIC EMISSION MONITORING TECHNIQUES</u>	7
BACKGROUND	7
EXPERIMENTAL SETUP	8
SOURCES OF AE	8
MONOLITHIC PIEZOELECTRICS	9
MULTILAYER PIEZOELECTRIC	12
CONCLUSIONS	18
<u>TECHNIQUE 2: THE USE OF OPTICAL IMAGING AS A METHOD OF DETECTING MICROCRACKING.</u>	19
BACKGROUND	19
EXPERIMENTAL MEASUREMENTS	21
MICROCRACK DETECTION	22
CONCLUSIONS	23
<u>TECHNIQUE 3: THERMAL IMAGING MONITORING TECHNIQUES</u>	24
BACKGROUND	24
PRINCIPLE OF OPERATION	24
EXPERIMENTAL MEASUREMENTS	25
CONCLUSIONS	27
<u>REFERENCES</u>	28

Aims

The aim of this work package is to investigate the feasibility of developing robust real-time evaluation (condition monitoring) techniques for the assessment of fatigue of electroceramic materials. The evaluation was performed on those piezoelectric materials which were characterised in the earlier milestones of CAM7 project. The fatigue of PZT materials can then be assessed using on-line monitoring techniques, which is considered commercially very relevant.

Objectives

The three following potential techniques have been examined. For each technique, the test materials were either virgin materials or were subjected to prolonged periods of representative electrical and mechanical stressing histories.

Technique 1: acoustic emission generated either by the spontaneous release of sound following domain motion or domain switching or by the growth and interaction of nascent flaws in the materials as it is exposed to representative electrical and mechanical stressing.

Technique 2: the change in the thermal signature of the material with time as visualised by a thermal imaging camera. Here representative piezoelectric multilayer actuators have been mechanically damaged in an effort to demonstrate the potential of this technique.

Technique 3: the use of optical imaging as a method of detecting microcracking in the material. This work has been conducted by the European SMT project EC-ACTUATE partner IMMG, based in Athens, Greece. This group have developed an imaging technique that is able to discern the formation of micron sized cracks from a distance of many tens of centimetres.

Executive Summary

This report aims to bring together the results from the various techniques developed to evaluate the real-time evaluation of degradation of piezoelectric materials. The work covers the measurements performed on piezoelectric samples and deals with the issues of degradation monitoring of these materials.

The electrical and mechanical fatigue exhibited by many piezoelectric and ferroelectric materials continues to limit their applications as actuators and many types of sensors and other functional devices. The monitoring, either in-line, on-line or off-line, of such materials represents a major technical challenge for industry. If methods can be developed such that unambiguous monitoring of degradation or performance may be made, then considerable savings, both economic and environmental, can be realised.

Techniques

The use of acoustic emission (AE) generation and detection within piezoelectric materials has been successfully demonstrated as a means of assessing internal physical processes occurring in multilayer materials and monolithic materials when driven at high electrical fields.

Experiments on monolithics have confirmed some of the findings reported in the literature of acoustic emission in piezoelectrics. Firstly, it is essential that a full understanding of the experimental set-up is developed and the possible sources of emission in the sample determined. It has also been found that it is necessary to include a resistor in the samples' high voltage connection to minimise high frequency noise. In soft piezoelectric material there seems to be a threshold below which no acoustic emission occurs, and the acoustic emission occurs mainly on a rising applied field. The acoustic emission is asymmetric between the positive and negative going signal, most likely due to the poled nature of the material. The application of a *unipolar* rather than a *bipolar* field almost completely halts acoustic emission. The effect of mechanical fatigue on the acoustic emission is to reduce the AE output, suggesting that there is little significant mechanical damage in the form of cracks, and that ferroelectric switching is being diminished.

It has been shown, that in multilayer actuators, the acoustic emission signal intensity depends on both the sample heating and the degradation of the multilayer. At present, it is difficult to deconvolute these two contributions. Further experimentation must concentrate on using lower voltages and reduced frequencies which will act to minimise sample heating. It will also be necessary to perform the measurements over a longer duration, as the initial period, where the degradation rate is expected to be fast, is difficult to characterise as the sample is 'settling in'.

The direct observation of microcracks within materials or devices using the system at IMMAG, Greece has been investigated. Optical imaging techniques are potentially capable of detecting micro-cracks, greater than 20-30 μm in piezoelectric ceramics. However, since the electroceramics are brittle materials and the crack size must be comparatively large in order to be detected, this optical imaging method can not be considered a sensitive technique to detect early stages of degradation.

The use of thermal imaging of piezoelectric multilayers has been shown to detect the thermal gradient caused by the combination of the self heating and the cooling of the device by the

ambient air. If damage within multilayer actuators is typified by interlaminar or electrode failure then it may be possible that 'hot-spots' may be caused by localised heating caused by shorting electrodes or electrical discharges across the piezoelectric layers. However the thermal signature is dominated by the self heating throughout the thickness of the sample and the likelihood that a small 'hot-spot' in one layer will cause a change in the thermal gradient of the whole sample is small.

If the method is to be used to monitor degradation in piezoelectrics then a much more sensitive detection system will be needed, or perhaps the present methods could be used on much thinner devices, perhaps thick or thin film devices.

Monitoring Degradation in Piezoelectric Materials

The electrical and mechanical fatigue exhibited by many piezoelectric and ferroelectric materials continues to limit their applications as actuators and many types of sensors and other functional devices.

Fatigue may be described by a decay in the polarisation and coercive field, associated with increasing electrical and/or mechanical stressing cycles. The attempts to explain the mechanisms for fatigue in these materials have been based on intrinsic or extrinsic phenomena including 90° and 180° domain wall motion and/or domain switching, domain pinning via space charge effects, surface deterioration via electric discharge within pores and, ultimately, the generation of microcracks in response to the very large strain accumulation within pores and imperfections. Thus, piezoelectric materials may undergo a change or decay in certain properties with time and mechanical and electrical stress. This has been discussed in a previous NPL report ¹.

Real-time evaluation of this degradation of properties is clearly of interest to many industries that utilise piezoelectric materials as either actuators or sensors. If the functionality of real materials and devices could be measured in real-time then this information could be exactly correlated with the driving conditions at the time any changes or deterioration was monitored. Even 'off-line' monitoring could yield information that would be of value to materials and device development. It is important that the technique is robust, reliable, relatively quick and simple to perform and of low cost if industry is to adopt it. Such a list of requirements is formidable indeed, and the work described in this report attempts only to describe the advantages of one technique over another. However, it may be possible for industry to rapidly gain some insight into the various merits and limitations of the techniques and how they may be adopted or adapted to suit their own needs.

A review of the literature reveals the dominance of *certain* 'real-time' techniques which have been used to monitor the degradation of piezoelectric materials. Most have been based on X-ray diffraction analysis measured in a clearly 'off-line' and certainly not 'real-time' scenario. However, this work has been useful in producing a wealth of scientific understanding of the phenomena associated with piezoelectric and ferroelectric degradation. Additionally, the degradation in thin ferroelectric films - developed for memory applications - has been extensively studied (for example see references in Ref.1). Acoustic emission techniques have been studied for many materials types and is a well known technique in the assessment of the mechanical properties of metals, ceramics and polymers. However, in the assessment of the degradation of piezoelectric properties AE has been of limited success. The technique has been thoroughly reviewed and investigated in succeeding sections of this report. Directly monitoring the formation of cracks - which may be described as one final outcome from serious degradation - has been investigated by our EC Standards Measurements and Testing Project partner IMMIG in section 2. Multilayer materials often degrade by an increasing level of inter or intra-layer delamination when subjected to high electrical or mechanical loading. Seen edge-on, the delamination may be viewed as the formation of micron-sized cracks. Such a system has been invented at IMMIG and demonstrated as a potential method of monitoring serious degradation in *real-time*. The final real-time monitoring technique is based on thermal imaging. Here, it is reasoned that if a piezoelectric material suffers damage through over-stressing then the thermal properties of that part of the material may be affected. 'Hot spots' or 'cold spots'

may be imaged using an infrared thermal camera, focused on the material, actuator or device in question. Alternatively, a material which has been degraded with time or stress may gradually become less and less effective as an actuation device, arising from a decay in fundamental piezoelectric properties or via a build-up of limiting defects within the material. The thermal properties may also become thus affected.

The following sections describe each technique in more detail. Results are shown and the validity of using the techniques as real-time monitoring evaluation is discussed.

Technique 1: Acoustic Emission Monitoring Techniques

Background

Ferroelectric ceramic materials normally contain a large number of sub-micron sized domains with symmetry linked to the materials crystal structure. If the material is poled (d.c. electrical field in excess of coercive field) then these domains may align themselves in the direction of the poling electrical gradient. This then produces a material with a poled domain structure and thus the ferroelectric becomes piezoelectric and pyroelectric. The degradation of such materials may be manifest as loss in piezoelectric performance, permittivity, change in loss factor and many other parameters. It has been shown that fatigue may be linked to various micro-domain and macro-domain changes, such as, domain wall motion and pinning of domain walls at impurities.

The fatigue performance may be monitored using various techniques, including X-ray diffraction^{2,3,4}, strain gauge⁵ and Acoustic Emission^{6,7,8,9,10,11,12,13,14,15,16,17}. The XRD and strain gauge techniques monitor changes in strain and crystallographic texture indicating a switching of 90° domains. The AE technique has been studied since the early 70's and used to describe the microscopic modifications and damage in ferroelectric materials and devices¹⁸.

Generally, the AE studies have shown that acoustic emission is generated through a sequence of events including domain switching, the paraelectric to ferroelectric phase transition and finally microcracking¹⁵. Additionally, electric discharges can also contribute to the AE spectrum in some materials¹⁸. Acoustic Emission is considered a useful tool for the investigation of fundamental domain dynamics in ferroelectric materials. In the investigation by Choi et al.¹⁵ two different La modified PbTiO₃ ceramics were used to be able to determine differences in AE spectra produced following 90° and 180° domain switching events. Following an analysis of the energy distributions, Choi demonstrated; that the AE signal could adequately discriminate 90° and 180° domain switching, that the AE signal from 90° events was significantly higher than that corresponding to 180° events and that the AE of 90° switching was dominated by burst emissions whilst that from 180° switching was dominated by continuous emission. Acoustic emission of thermally shocked piezoelectric materials revealed that even in the absence of switching electric fields substantial emission was recorded and was considered a useful tool for the assessment of reliability of ceramic actuators⁹.

A set-up able to distinguish the amplitude, rise-time, total signal length, event energy, the entire transient signal and its Fourier transform and the localisation of the AE event has been described by Lupascu¹⁸. For soft PZT materials, a distinct threshold in electrical field for acoustic emission was observed. Additionally (since AE is not generated through simple domain wall motion - unless it is hindered or blocked in some microscopic manner) the large strains induced via domain wall motion in soft materials did not produce any AE until very high strain levels were recorded. Subsequent polarisation switching generated AE even at low fields. Conversely, hard materials exhibit AE continuously during the entire hysteretic cycle with no periods of loss of AE signal relating to minimal material deformation. Interestingly, in this paper, significance is made of the drive electronics circuitry in masking or mimicking domain dynamics in ferroelectric materials. Attempts to determine whether the measured AE signals originated from either the driving equipment or internally from the ferroelectric material were described. If the signal decreased in intensity with time then it is likely to be a real material property rather than an artefact from the electronic noise of the amplifier.

It is the purpose of this section to investigate the feasibility of using acoustic emission as a tool to monitor degradation in electroceramic actuators. To this extent the next sections describe the experimental set-up and main results achieved.

Experimental Setup

The Acoustic Emission measurements were performed using a Physical Acoustics Corporation MISTRAS 2001 PC based DSP data acquisition card. The system can handle two AE sensors and two additional signal inputs which in these experiments were used to record the temperature and the voltage applied to the piezoelectric sample.

Using the incoming AE signal the MISTRAS software can make measurements of the waveform, dependent on several interactive parameter settings used to identify an acoustic event or 'hit'. The most important parameter to determine is the threshold, below which it is assumed to be noise, and there are several other parameters that enable the correct identification of the event. The lower limits of these parameters define the fastest single event that can be measured and recorded using this system, and is of the order 20 microseconds.

The AE signal was simultaneously captured on a 20MHz Gould 840 digital storage oscilloscope in order to examine the AE signal in more detail.

Sources of AE

It is difficult to come to any coherent conclusions about Acoustic Emission behaviour in piezoelectric materials as many published experiments seem to disagree. One possible reason for this is that it is easy to misinterpret some experimental artefact as true acoustic emission from the sample under test. The following section attempts to list the possible ways that an acoustic emission 'hit' will be produced, and gives a rationale for isolating spurious hits.

It is always preferable to be able to see the raw AE signal on a fast triggering oscilloscope to give some idea of the amplitude and frequency of the emission. As a starting point there should be no AE detected when the sample is merely attached to the system electronically, and when there is **no** applied field. There is often, however, some emission that is caused by the relaxation of stresses - mechanically and thermally induced - immediately after the sample is placed in the experimental set-up. These should quickly die out until there is virtually no AE. If at this point there is still consistent AE output possible causes are: through external mechanical vibration such as fans on electronic equipment; through electrical interference, equipment that switches power on and off such as soldering irons. Even if the sample holder is reasonably mechanically isolated there is always the possibility that mechanical vibration can be transferred through the sensor cable.

The next test to carry out is to switch on the power supply - usually high voltage that is to apply the field to the piezoelectric - with all the connections made to the sample but with the driving voltage set to zero. There should be no increase in signal output from the AE sensor as there should be no driving signal. If changes are observed, however, possible causes could be electronic interference from the high voltage amplifier to the sensor electronics. However, in this work and in others¹⁹ the cause is due to high frequency noise from the amplifier output. If the HV output is examined on an oscilloscope, a considerable amount of high frequency noise is found, which causes the sample to resonate leading to acoustic emission. The sample itself is

a capacitor and so by the simple addition of a resistor in series with the sample, a smoothing circuit is built, which filters out this high frequency noise and cuts out the spurious acoustic emission.

Once this condition is reached the driving voltages can be applied to the sample. However, even now there are sources of acoustic emission that do not arise from the material. Electrical interference from an amplifier that arises under loaded conditions is almost impossible to sort out, but usually if the signal changes with time - or some other independent modification of the sample - then it is true acoustic emission.

Sample acoustic emission could be caused by domain switching, cracking in the material, or high voltage breakdown within pores in the material. It is also possible to get discharge around or between poorly made contacts which are an experimental artefact. Similarly, if the sensor is not firmly attached, the sensor could physically 'bounce' around on the sample if relatively high driving frequencies are used. Although in some of the earlier experiments in this work the sensor was attached to the sample using a soft wax, this was later reinforced by mechanical clamping. Figure 1 shows the sample set up for measuring AE from a piezoelectric multilayer, where the temperature of the multilayer is monitored with a thermocouple situated below the sample. The clamping arrangement for examining monolithic piezoelectrics was similar except the sample was immersed in a silicone oil to prevent electrical discharge.

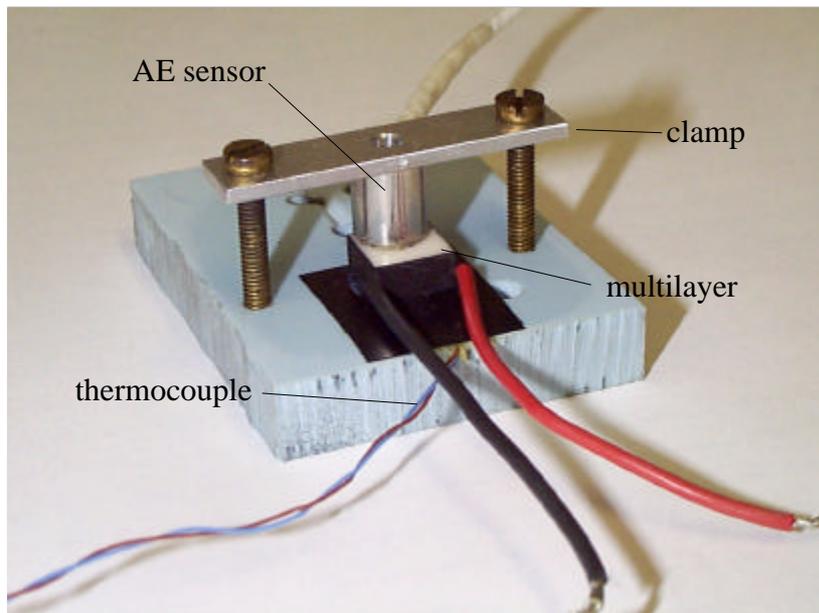


Figure 1: Photograph of AE / Actuator setup. Temperature was measured using a thermocouple beneath the device.

If the AE signal is examined on an oscilloscope there is often a large component of a single frequency present in the waveform. The signals were at first thought to be due to individual acoustic emission events, but as the frequency was found to be in the range 100-300kHz this was thought unlikely. The most likely cause is resonance in either the sample, the sensor or some combination of the two. As the AE sensor is also a piezoelectric the resonance behaviour of the sample and sensor can be measured

using an impedance analyser. At first the measured frequencies did not correspond exactly to either the sensor or the sample, but of course the clamping set-up in the AE experiments changes the resonance behaviour. If the impedance sweep was performed on the sample and the sensor in situ it was possible to pinpoint the dominant resonant component.

Monolithic Piezoelectrics

In order to get some confidence in the experimental set-up several experiments were performed which attempted to repeat some of the results determined in the literature. Much of the work reported in the literature has examined switching behaviour at high electrical fields and necessitate a covering of oil to prevent breakdown in air. The material chosen for study was a soft 5A type material. Lupascu¹⁸ has reported that this material shows a lessening of the acoustic emission around 1.2-1.6kV/mm, and has attributed this to the stress free movement of domains in a material with few pinning sites.

Again, most reported work has tended to concentrate on very slow electrical driving field ramp rates so that the chances of detecting amplitude-dependant acoustic emission is increased. The ramp rates used in the monolithic experiments was 20mHz with amplitudes of up to 6kV/mm p-p. Figure 2 shows a plot of acoustic emission energy (y-axis) against the amplitude of the driving field (x-axis). There would seem to be a threshold of around 1.4kV/mm before any significant acoustic emission occurs and the levels increase with increasing amplitude. There does not seem to be any evidence of the quiet region found by Lupascu in our 5A samples, although this could be because the materials in this study were sourced from a different manufacturer. Figure 2 also shows there is a difference between the positive and negative acoustic emission. This asymmetry in the samples was also seen for the actuator materials, however it was not possible to quantify it or develop an understanding on why this should be. It was also noticed on the oscilloscope signal that the majority of the emission occurred on a rising signal, either positive or negative, and there was little emission as the driving field returned to zero.

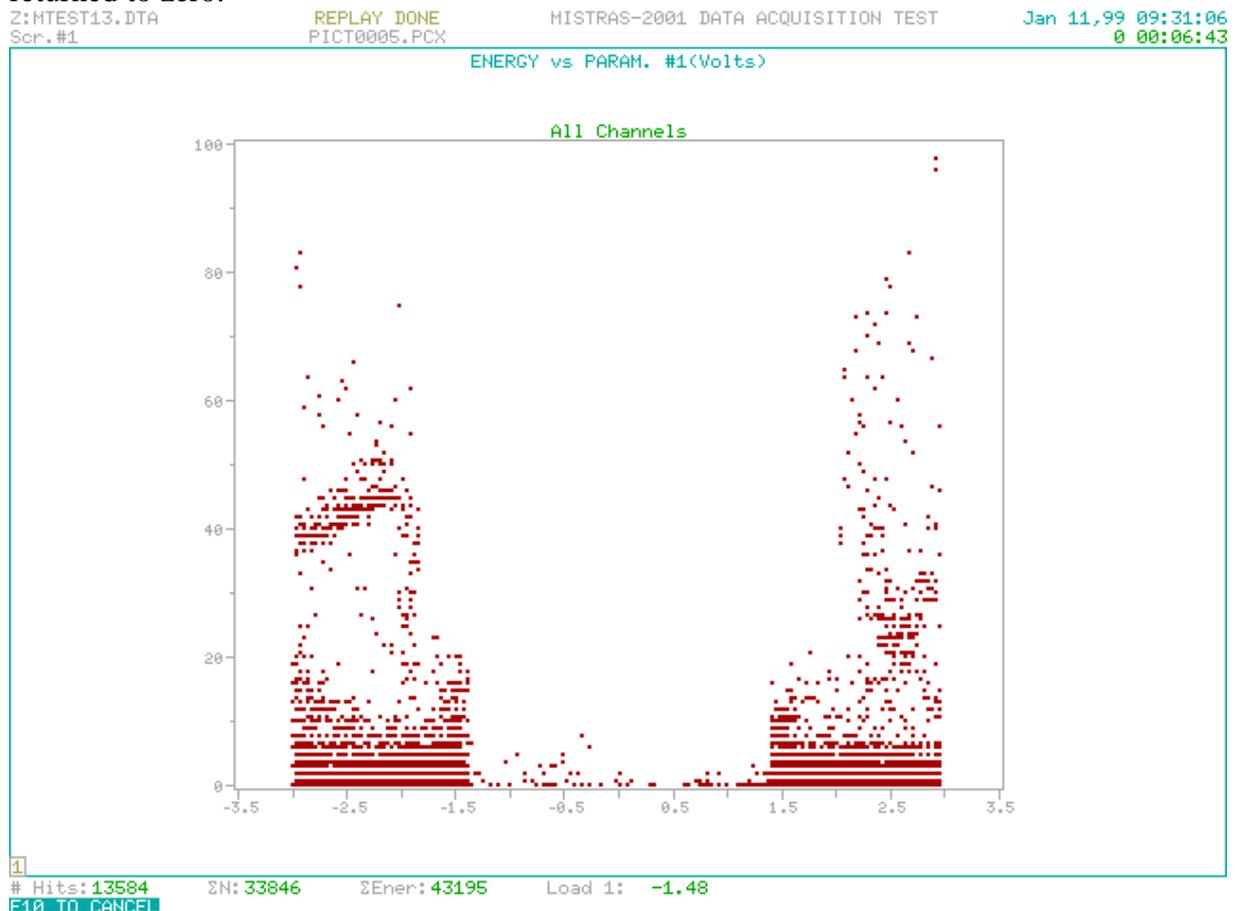


Figure 2: Acoustic emission energy against driving field (PARAM. #1). Units y-dB, x-kV/mm

Aburatami and Uchino¹⁹, who first reported the addition of an RC network in the high voltage set-up, found a difference between unipolar and bipolar driving fields. They found that the acoustic emission was virtually eliminated under unipolar drive and suggested that this was because the emission was mainly due to 180 degree switching, rather than non 180 degree events. They also found that the acoustic emission was greatly diminished on unipolar drive, and additionally that the signal gradually decreased on changing from bipolar to unipolar. Figure 3 shows the number of AE counts decreasing with time after switching to unipolar drive. The first cycle is obviously switching domains that were switched in the bipolar fields, however it takes several unipolar cycles to switch all these back.

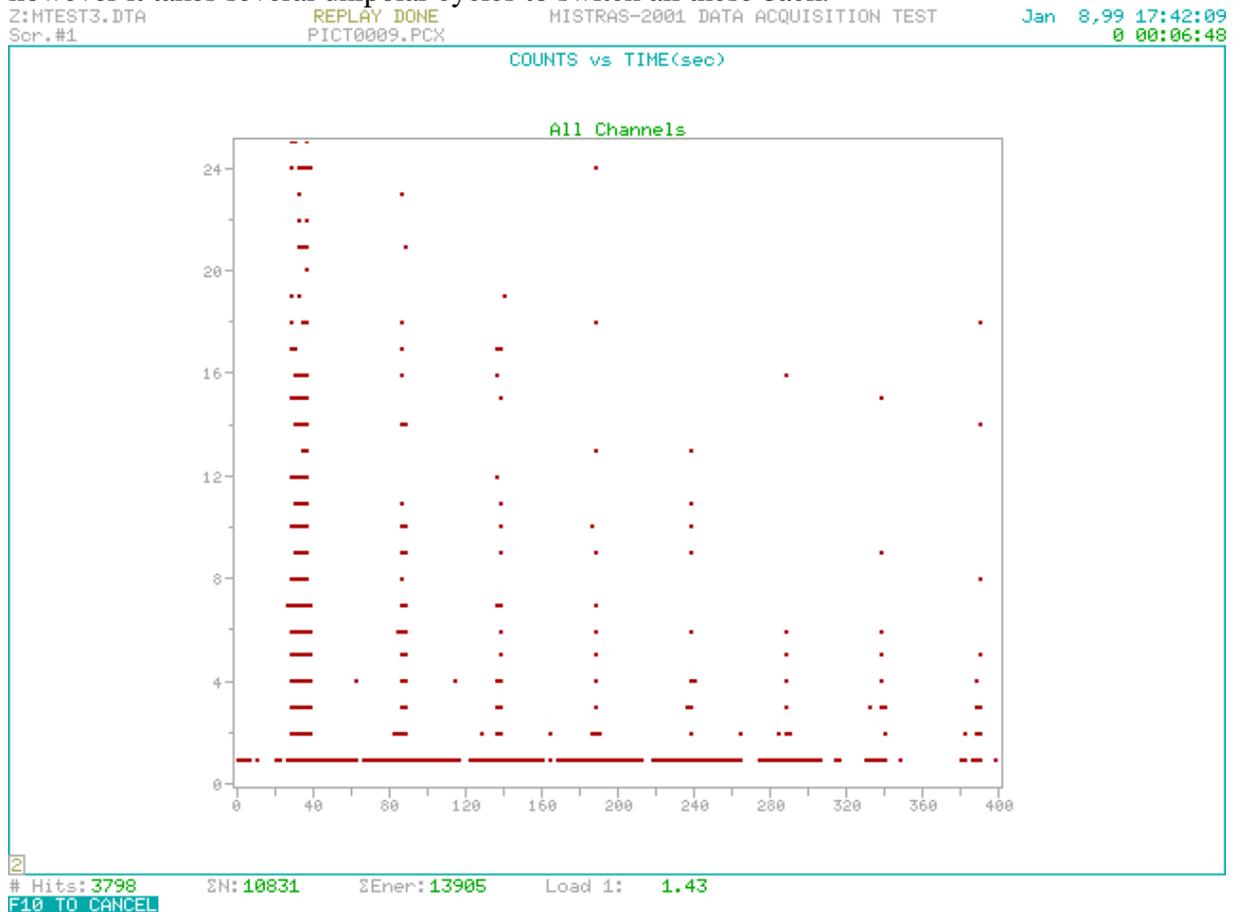


Figure 3: Acoustic emission counts vs time for a sample that has been switched from bipolar to unipolar drive showing a diminishing signal with time

In order to investigate whether there was any change in AE as piezoelectrics degrade, some samples that had been mechanically fatigued were studied. The acoustic emission in the fatigued samples tended to be reduced as compared with the as received samples. It was not possible to measure the AE whilst the samples were mechanically fatiguing because the fatiguing equipment create too much noise. Figure 4 shows the AE output for a sample that has seen 10^4 cycles of 30MPa cyclic compressive loading, and compared with Figure 2 is greatly reduced. This would appear to conflict with evidence that shows that mechanical damage in the form of cracks greatly increases AE. However it may be that the fatigue has not been sufficient to produce cracking, and it is the 180 degree switching which is inhibited.

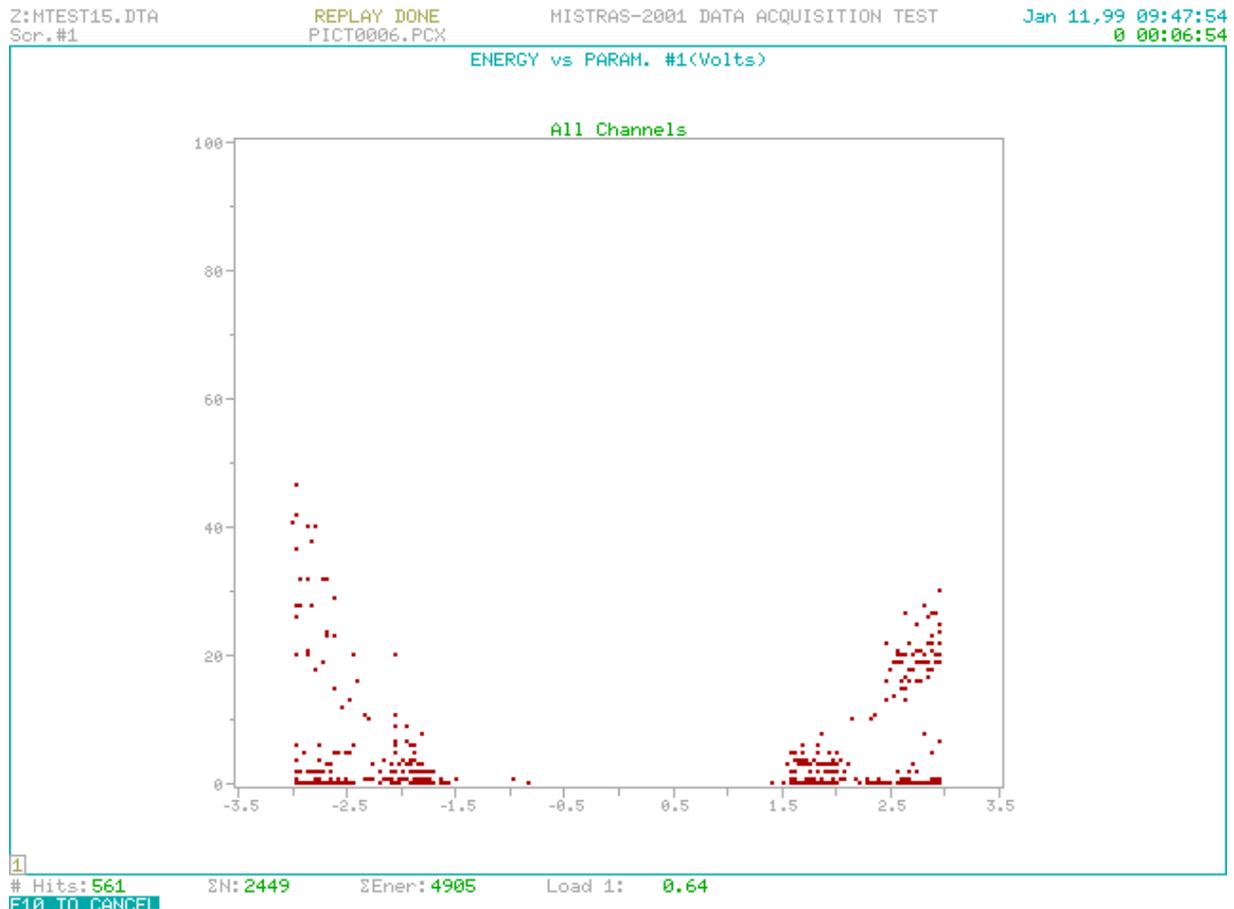


Figure 4: Acoustic emission energy vs driving field for a sample that has seen 10^4 cycles of 30MPa cyclic loading

Multilayer Piezoelectric

Acoustic emission experiments were also performed on multilayer piezoelectrics, 10mm by 10mm by 3mm thick made from a soft piezoelectric. Because of the large capacitance of these devices the driving voltages and frequencies were limited, and were kept near the manufacturers recommended driving voltage. The aim of these experiments was to measure acoustic emission in these devices, and to see if the signal changes with repeated exposure to the driving field.

Using a sine wave drive, acoustic events were almost constantly occurring, and although it was possible to see the relationship between the driving voltage and the acoustic emission, it proved impossible to make any meaningful measurements using the MISTRAS software. With a square wave drive discrete bursts of acoustic emission were produced every time the voltage levels switched, and so it was possible to make measurements of these pulses. The pulses produced with the square wave drive were also much louder than those produced from the sine wave drive, in fact, so as not to saturate the system the preamplifier gain had to be set to 20dB, from an initial setting of 60dB which was needed for reasonable results using the sine wave drive. The major problem using the square wave drive was that the amplifier system could not maintain a perfect square wave, due to the high slew rates and the large capacitance of the samples. Using a sine wave drive at voltages of 150 volts it was possible to go to 20Hz

without excessive distortion of the input drive. Using the square wave the distortion was apparent even at 0.1Hz (as expected from the transient drive of a square wave). However, the input wave form was stable enough to trigger the DSO and capture the acoustic emission consistently.

Figure 5 is the DSO output showing the AE signal, applied voltage and the current measured as the voltage drop across a 100 ohm resistor in series with the sample.

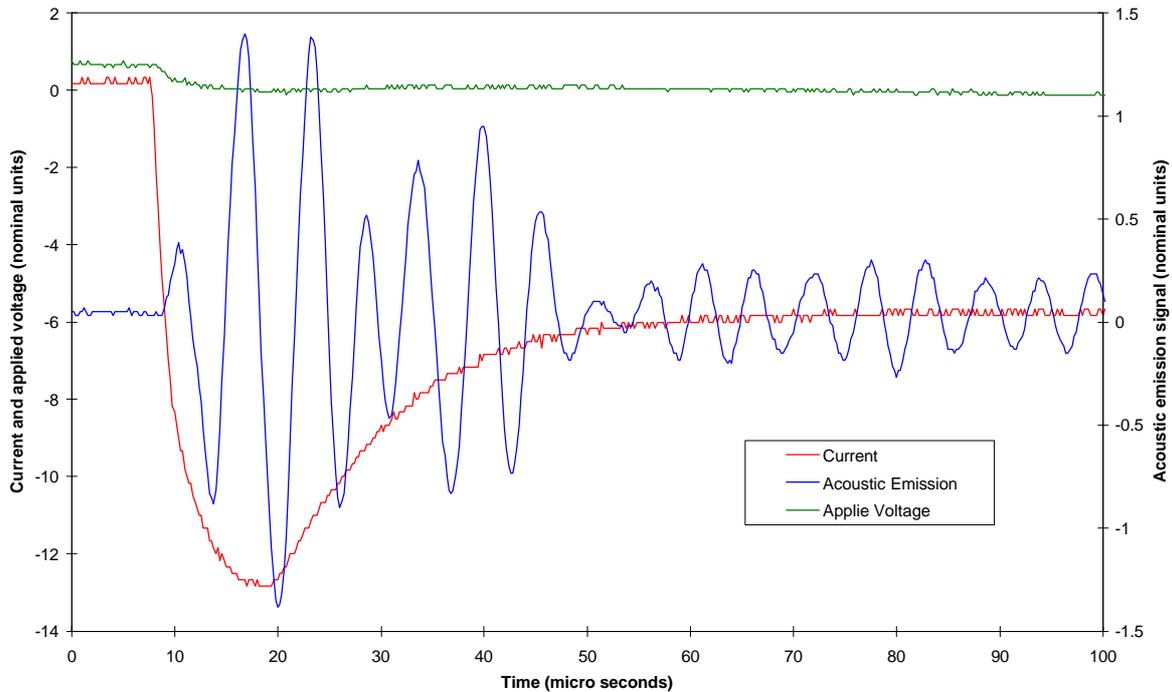


Figure 5: DSO output showing the AE signal, applied voltage and the current measured as the voltage drop across a 100 ohm resistor in series with the sample

The MISTRAS software was set to record the amplitude, energy, counts and duration of each hit, and also record the applied voltage and temperature at the time of the hit. It is possible to record many more parameters, however this then increases the data storage for each scan. Figure 6 shows the a graph of energy against cycles, for a sample that had been left cycling for many cycles before these results were recorded. Interestingly they show there are two pulses with different energies, one at 78 and the other at 87. In fact these differences are due to the sense of the voltage change. If the energy of the pulse is plotted against the applied voltage that the pulse occurs (Figure 7), it can be seen that the higher energy pulse occurs on the positive side. This figure also shows that adding an offset to the applied voltage changes both the energy and applied voltage of the events, however the energy for the positive side is consistently higher. It is interesting to note that the direction of the offset also produces a non symmetric change in behaviour. This non symmetric behaviour is similar to that seen in the monolithic samples, and is probably a consequence of the poled nature of the multilayer.

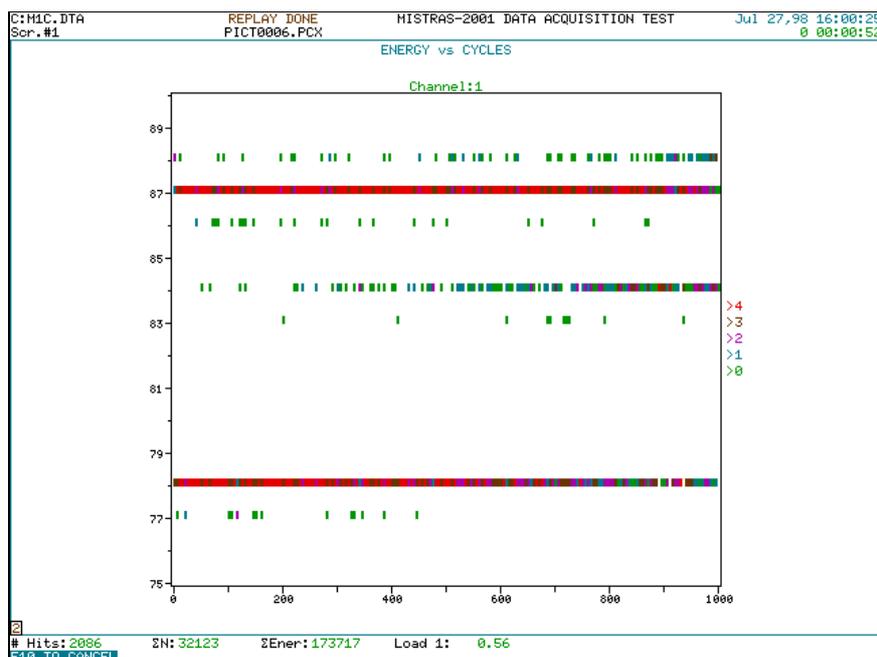


Figure 6: Acoustic Emission energy against number of cycles for sample excited with 300 volt p-p square wave at 20Hz. Sample had reached thermal equilibrium.

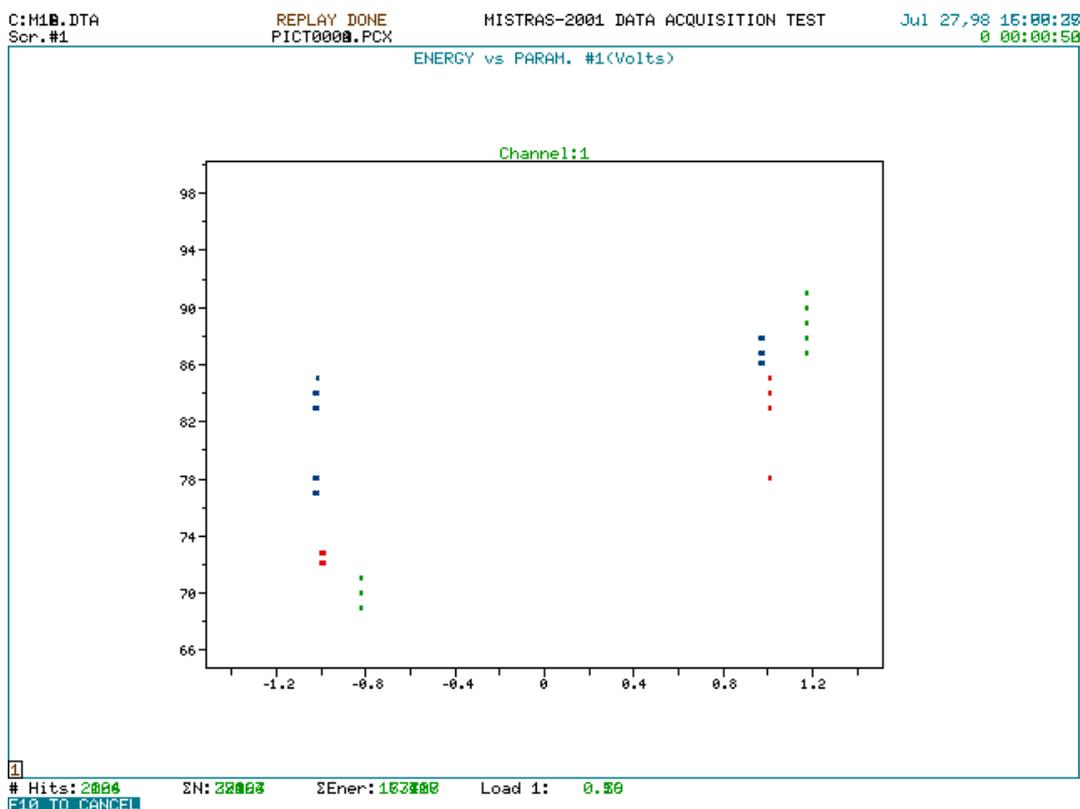


Figure 7: Acoustic emission energy against applied voltage, showing the effect of adding an offset to the applied voltage. The x axis shows the voltage monitor which is 1/150 of the applied voltage.

In an attempt to produce large amounts of acoustic signal and introduce sample damage the voltage and frequencies used are higher than the rating of the device, and consequently the sample heats up considerably. If the acoustic monitoring is started when the sample is at ambient then there is a gradual increase in the energy of the pulses which mirrors the sample temperature, and during this period there is no discernible difference between the positive and negative pulses. Only after 30 or so seconds does the energy signal spilt into two distinct energies (Figure 8). Figure 9 shows the corresponding DSO captures for the first and the 40th second after starting. This shows the amplitude and duration of the acoustic event has increased, i.e. the energy has increased, but there is very little difference in the current trace. It is obvious that the increased acoustic emission is linked with the change in temperature of the sample but whether it is caused directly by this or as a manifestation of the same phenomena is not clear. In all cases the energy of the acoustic emission and the temperature increased at the beginning of the test. However, as the temperature stabilised, the acoustic emission energy tended to decrease.

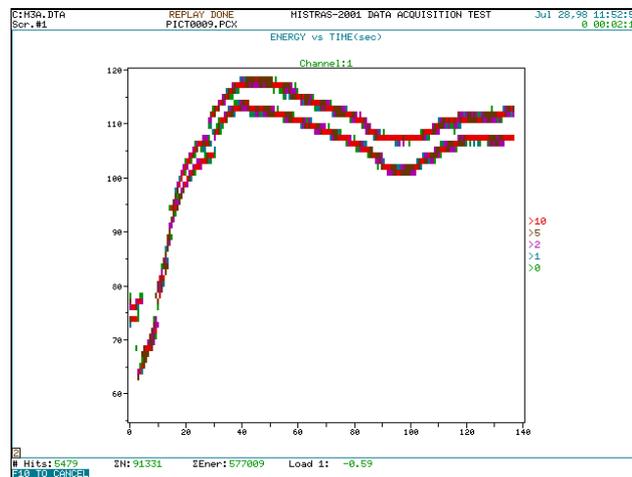


Figure 8: Acoustic emission energy against time after first applying the voltage to the sample

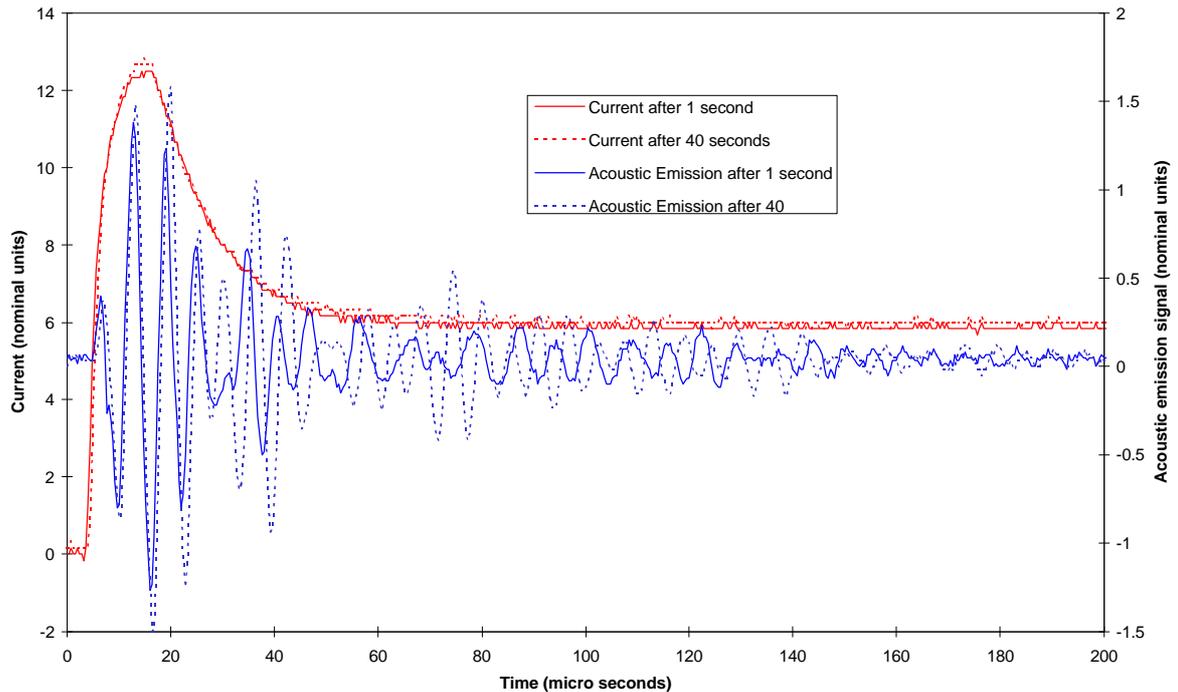


Figure 9: AE signal 1 second and 40 seconds after applying the excitation field to a ‘used’ sample.

Changing the frequency of the square wave had no noticeable effect on the acoustic emission as monitored on the DSO and it is expected that there will be a decrease in the heating effect although this was not measured.

For most of the preceding experiments, in order to investigate the sensitivity of the test method, a sample was used that had seen many electrical loading cycles, i.e. where little degradation was expected to take place. Figure 10 shows the acoustic emission of the first 240 seconds of exposure of a virgin sample. The acoustic energy changes greatly over this period, but initially the energy decreases as the sample heats up, which is contrary to the findings on ‘used’ samples. Also, there is a change in the voltage at which the pulses occur on the negative side (Figure 11). Initially the pulses start at around -0.9 volts, and then, as the pulses reduce in energy, the voltage at which they occur decreases to -1.1 volts. Over this time the voltage level at which the positive pulses occur stays relatively constant. Again this non symmetrical behaviour is due to the poled nature of the sample, and there is some degree of depoling due to the high driving fields. Near the end of this experiment the AE sensor came off the sample, so some of the signal variations could be due to the movement on the AE sensor on the sample. Figure 12 shows the thermocouple voltage from a K type thermocouple attached to the sample, and it can be seen that even after 240 seconds the sample temperature is still increasing.

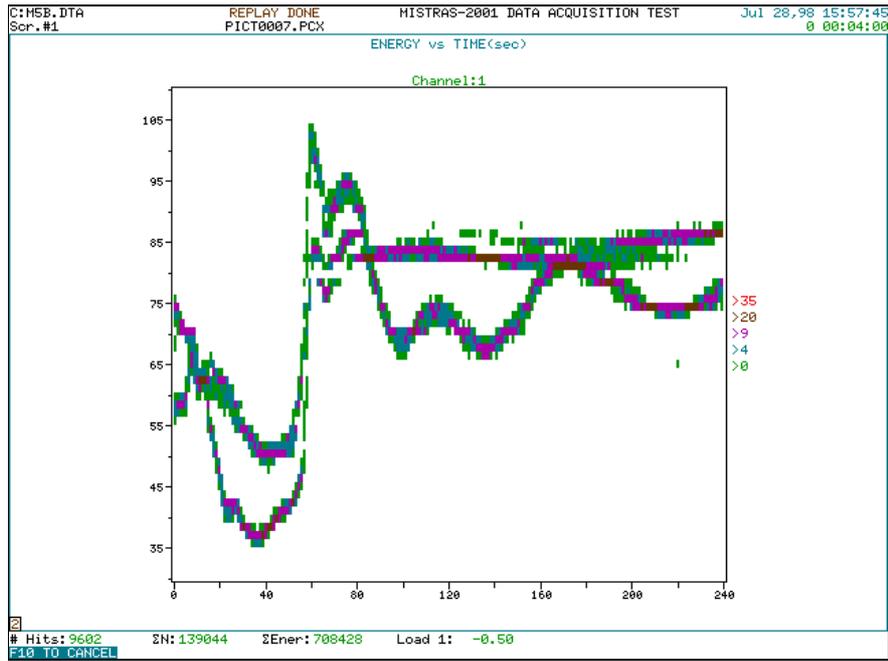


Figure 10: Acoustic emission energy against time after cycling a virgin sample at 20Hz with a 300 volt p-p square wave.

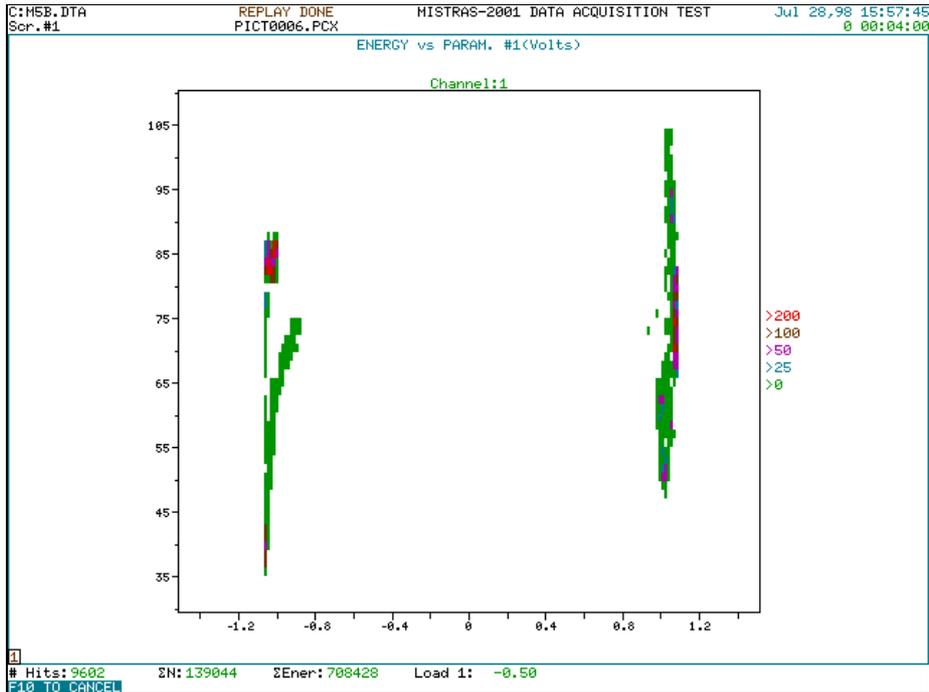


Figure 11: Acoustic emission energy against applied field for sample in Figure 10.

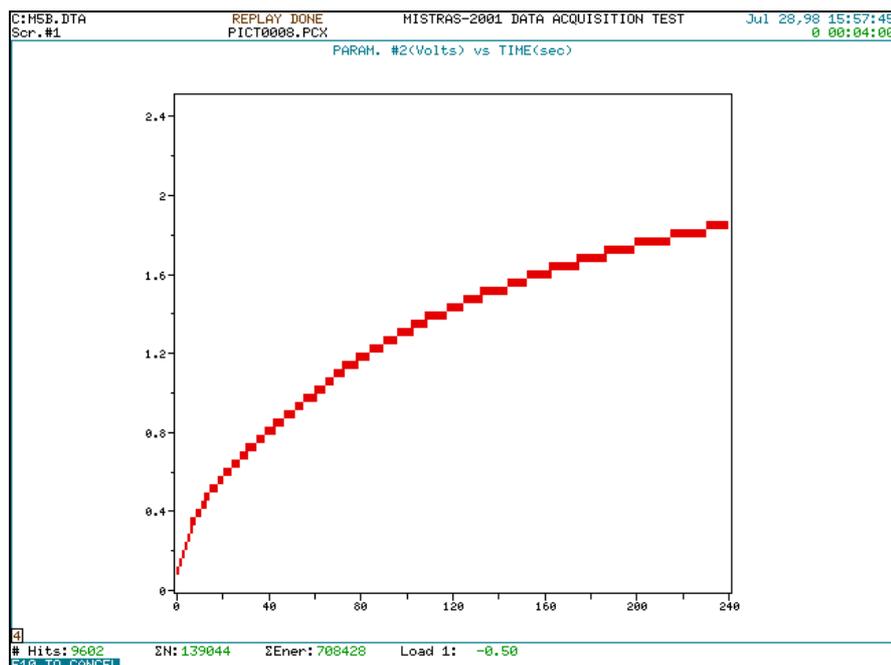


Figure 12: Thermocouple voltage (mv) from a K-type thermocouple attached to the sample.

Conclusions

Experiments on monolithics have confirmed some of the findings reported in the literature of acoustic emission in piezoelectrics. Firstly, it is essential that a full understanding of the experimental set-up is developed and the possible sources of emission in the sample determined. It has also been found that it is necessary to include a resistor in the samples' high voltage connection to minimise high frequency noise. In soft piezoelectric material there seems to be a threshold below which no acoustic emission occurs, and the acoustic emission occurs mainly on a rising applied field. The acoustic emission is asymmetric between the positive and negative going signal, most likely due to the poled nature of the material. The application of a *unipolar* rather than a *bipolar* field almost completely halts acoustic emission. The effect of mechanical fatigue on the acoustic emission is to reduce the AE output, suggesting that there is little significant mechanical damage in the form of cracks, and that ferroelectric switching is being diminished.

It has been shown, that in multilayer actuators, the acoustic emission signal intensity depends on both the sample heating and the degradation of the multilayer. At present, it is difficult to deconvolute these two contributions. Further experimentation must concentrate on using lower voltages and reduced frequencies which will act to minimise sample heating. It will also be necessary to perform the measurements over a longer duration, as the initial period, where the degradation rate is expected to be fast, is difficult to characterise as the sample is 'settling in'.

Technique 2: The use of optical imaging as a method of detecting microcracking.

Contribution from Dr Paul Michelis, IMMIG, Greece²⁰.

Background

The Institute of Mechanics of Materials and Geostrucures (IMMG), based in Athens, Greece, has upgraded a long distance focusing microscope (20 cm) in order to detect and record microcracking activity within piezoelectric ceramic materials. The microscope can change position in all three directions X, Y, and Z with the use of three accurate micro-step motors

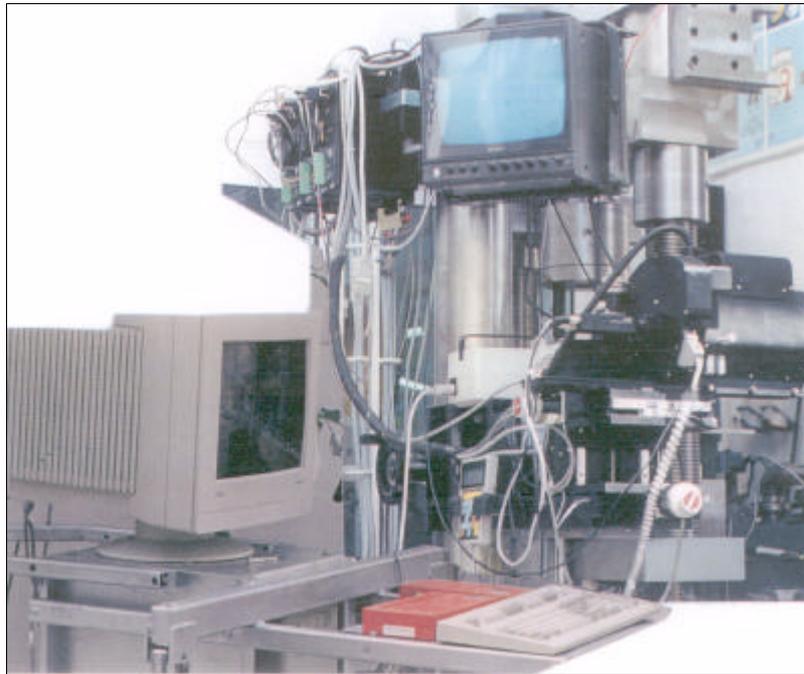


Figure 13: A long distance focusing microscope, a 3-axis controller and image hardware.

(Figure 13). A software package controls the microscope and the three motors.

The basic steps followed during imaging are the following:

- The user selects a point as a reference for the microscope co-ordinate system.
- The co-ordinates of each spot on the investigated surface are recorded. The user operates the software to get the X, Y, and Z measurements for each spot.
- A program stores all these measurements automatically in a data file. Additionally, the software stores an image of the area around the spot. This image describes the spot and is used to verify the accuracy of the measurement (Figure 14).
- During an experiment the software can measure the new co-ordinates of each spot, and store them in a data file. This can be repeated many times in order to get a significant number of measurements. The deformation is the difference between two successive measurements (i.e. $\Delta x = x_1 - x_2$).

This process has the following advantages over the current measuring methods :

- Gives accurate deformation measurements. The accuracy of these measurements

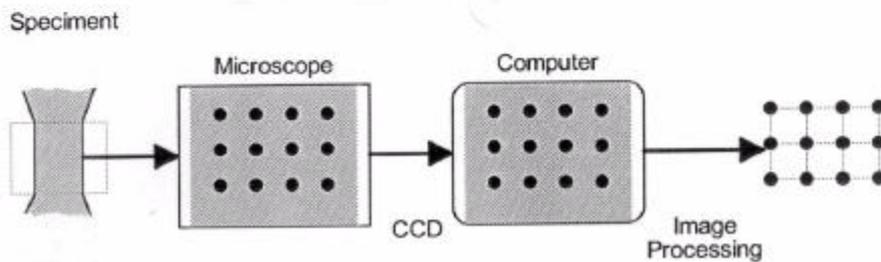
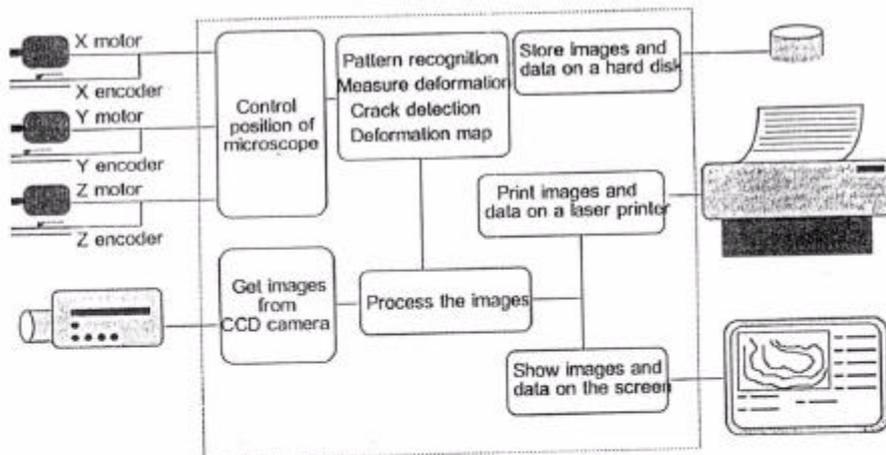
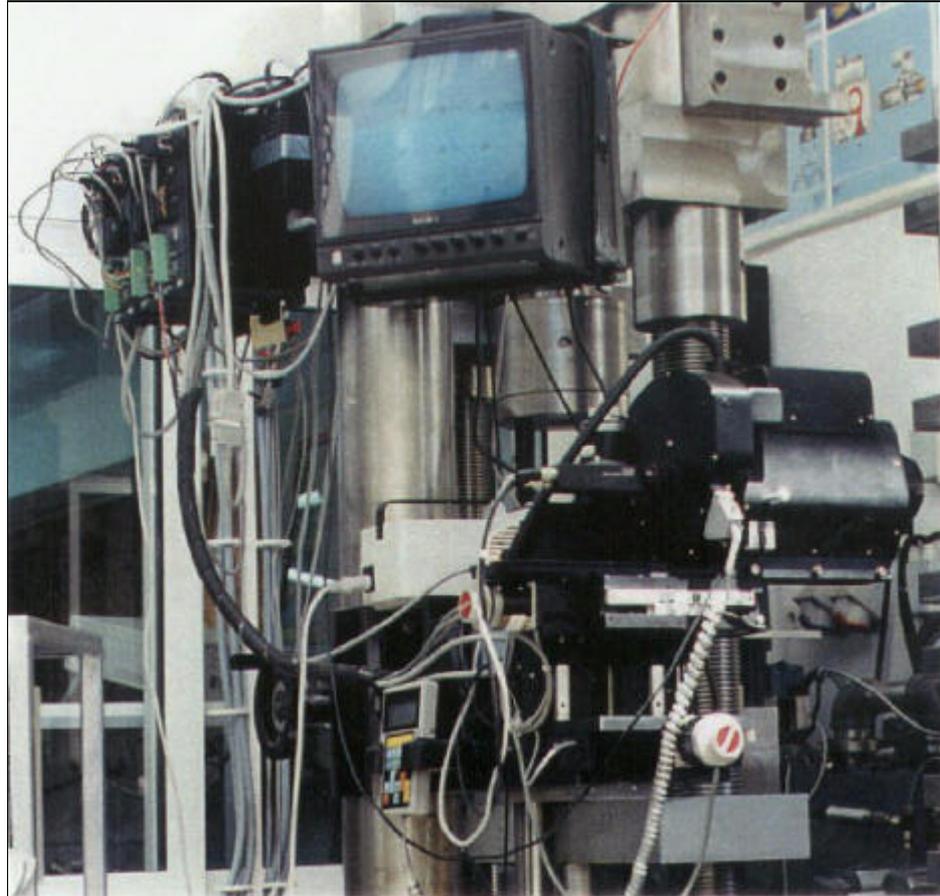


Figure 14: Optical in plane deformation measurement. The long distance observation microscope, the software package and the process flow chart.

can be $2\mu\text{m}$.

- The recording of images for each spot gives the ability to verify the measurements.
- The images can show interesting areas during the experiment.



Figure 15: Example of optical deformation measurement and microcracking activity investigation with the computer vision system.

Experimental Measurements

A number of image processing techniques can be applied to give detailed information about the specimen. Contrast enhancement can show more details on the image (Figure 15).

Edge and crack detection algorithms can automatically detect edges and cracks on the specimen. Pattern recognition techniques can provide automatic detection of the spots and can eliminate errors that are related with the user input.

Microcrack detection

A piezo-stack of ten PZT-4D rings was cyclically loaded in compression (25 MPa peak, 5 MPa minimum, Figure 16).

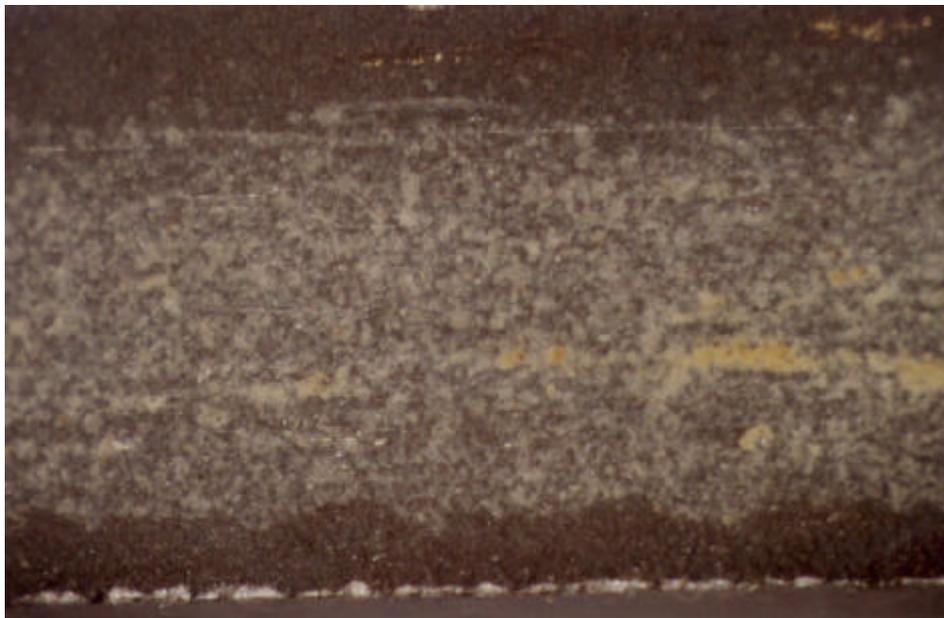


Figure 16: Micro photograph of the lateral surface of a PZT-4D ring.

High magnification images were taken every 10^6 cycles at three predetermined positions close to where the strain gauge rosettes were bonded. The gauges were used to provide information about the local deformation. Also the rosette grid was used as a grid of markers for the microscope. This procedure allowed a calibration of the optical deformation measuring system. The objective was to observe material degradation by detecting microcracking activity.

After several iterations new "spots" and "zones" were added in the previous microphotographs and analysed using a high resolution scanner (4096 pixels, 6144 steps/35 mm). The analysis revealed changes in the specimen surface.

A limitation of this technique is the small field of view associated with the long focal length camera. Although the imaging was focused on the high stressed area of the sample under investigation, one disk had cracked in an area away from those under observation.

Conclusions

- Optical imaging techniques are capable of detecting and recording micro-cracks, greater than 20 to 30 μm , in piezo-ceramic materials. The technique can follow the crack growth.
- In addition, optical imaging can provide local deformation information along the cracks and between markers (spots).
- The detailed electronic information produced for one image is extremely large (fills a complete CD) and therefore can not be easily analysed and efficiently compared with previous images. Presently, the system is not at the state of being able to determine image differences, although several edge-enhancement techniques have been used.
- The slight colour intensity "changes" observed can not be undoubtedly associated with real material property changes (for example development of micro-voids). Well defined microcracks or cracks were not observed. All the specimens failed suddenly.
- Since the electroceramics are brittle materials and the crack size must be comparatively "large" in order to be detected, the optical imaging should not be considered as a technique sensitive enough to detect degradation from its early stage.
- IMMAG will upgrade its imaging capabilities incorporating LabView hardware-software in the Questar microscope system. Laser imaging is also being considered.

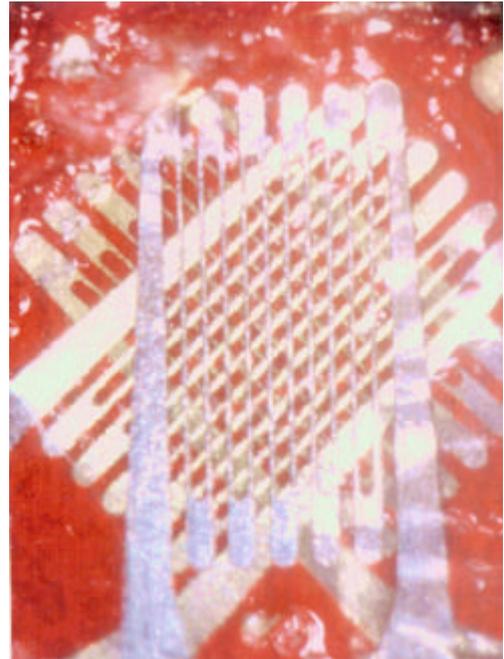


Figure 17: A rectangular strain gauge rosette viewed through the long distance focusing microscope. The specimen is a PZT-4D ring and the gauge grid dimensions are 1 x 0.7mm.

Technique 3: Thermal Imaging Monitoring Techniques

Background

In thermal imaging the infrared radiation emitted by surfaces at different temperatures is focused onto a sensitive plate in a similar manner to the formation of a photograph using visible light. Normally, the system is electronically controlled and a raster scan image is the result. Non destructive evaluation using infrared thermal imaging has been developed for the rapid on-line inspection of welds, coatings and composite structures²¹. For example, precise thermal NDE has been developed as a wide-area inspection tool to quantify structural damage within airframes and bridge decks²². Surface crack length measurement based on the use of thermal imaging systems have been developed²³ whereby an open crack can be distinguished from surrounding material due to its greater thermal radiance. It is these principles which have provided the impetus for this study of on-line monitoring using thermal imaging.

A review of the ferroelectrics literature has revealed that very little effort has been devoted to the study of degradation in ferroelectrics using thermal analysis. The reasons may have become clear during our study. The thermal signatures or differences between damaged and pristine parts of a multilayer ceramic actuator were not resolvable using the system at NPL. It is possible that thermal diffusion will always mask any slight aberration in absolute temperature within the device. Alternatively, the thermal camera was simply not able to resolve the differential temperature or spatially resolve the slight fluctuations that may be present in a damaged material.

Principle of Operation

The thermal imaging camera (NEC 2100) used in these experiments to measure the infra-red radiation given off by the object being measured, uses the well known relationships between infra-red radiation and temperature to derive a temperature measurement. The NEC 2100 uses the 8-13 μm wavelength-band to measure temperature.

The detector unit optionally scans the surface of the measured object sequentially from left to right and top to bottom for infra-red radiation by using mirrors (Figure 18). The infra-red energy obtained in this way is collected by an infra-red objective, and after chopping with a reference temperature source, is collected again by a relay lens and converted into electrical signals proportional to the incident energy using an HgCdTe detector which is cooled by liquid nitrogen. Focus is achieved by adjusting the focal position of the objective lens from the minimum of 20 cm to infinity.

Electronics in the control unit compensates for environmental variations in temperature using the temperature sensor in the detector unit. The signal is linearised to a signal proportional to temperature.

The system has a zoom capability up to 3x, an image size of 256 by 191 giving a spatial resolution at the relevant focus position of 0.5mm.

The measurements are clearly affected by the emissivity of the object being measured. As the purpose of the measurement was simply to ascertain if useful qualitative measurements could

be made, the emissivity setting for the instrument was left at 1.0 for the experiments carried out here.

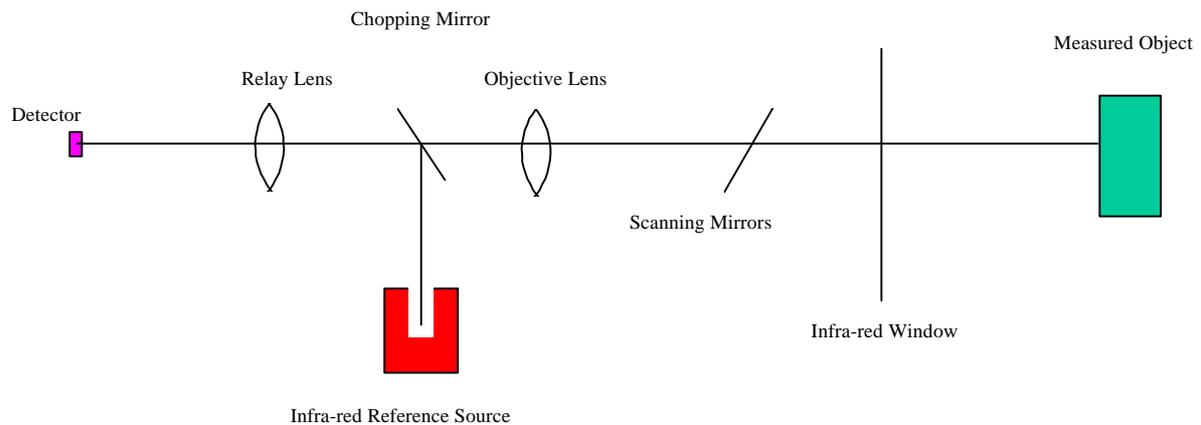


Figure 18 Schematic of detector

Experimental Measurements

The experimental sequence for on-line detection of degradation consisted of driving several multilayer actuators at double their maximum stated driving voltage. The surface of the square multilayer device was imaged using the IR thermal camera. The sample was rigidly affixed to a thermal conducting block of aluminium using adhesive tape, Figure 19. A voltage was applied to the device in the form of a positive and negative going sine wave of 150V pk/pk. This represented an overdrive of +33%, -100% with respect to the manufacturers data sheet. The devices were degraded over a period of time and re-inspected using the thermal imager and compared to the image of the pristine sample. It became clear that thermal imaging was unable to detect any changes in thermal radiance, at the thermal and spatial resolution afforded by the system. The movie file, shown in movie 1, is representative of a series of still images captured using the camera and subsequently integrated into a real time movie (avi windows compatible). Please double click the icon contained within the Movie placement below to start the show (pdf versions of this document only).

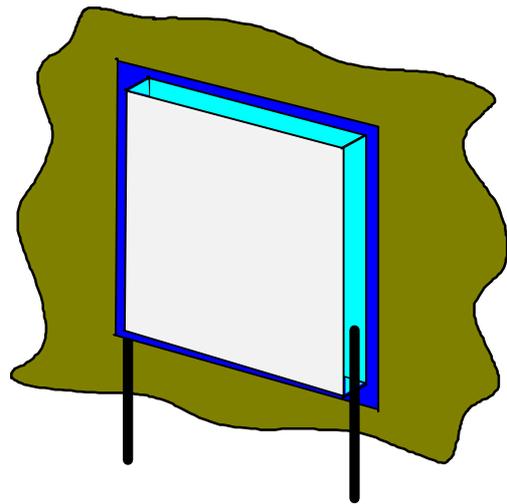


Figure 19: Thermal Imaging of multilayer actuator

In an effort to sustain real physical damage to the multilayer device, a piece of the front face was delaminated using mechanical means. The same multilayer was then inspected using thermal imaging, in an attempt to discern any thermal differences brought about by the serious surface damage. The sample is shown in Figure 20, and the resultant thermal image at maximum applied voltage is shown in Figure 21. Clearly, even with substantial damage imposed upon the surface of the actuator the thermal camera is not able to pick up any differences in thermal signature. The thermal image clearly shows the thermal gradient caused by the combination of the



piezo2.AVI

Movie 1: AVI clip of thermal image for driven multilayer actuator.

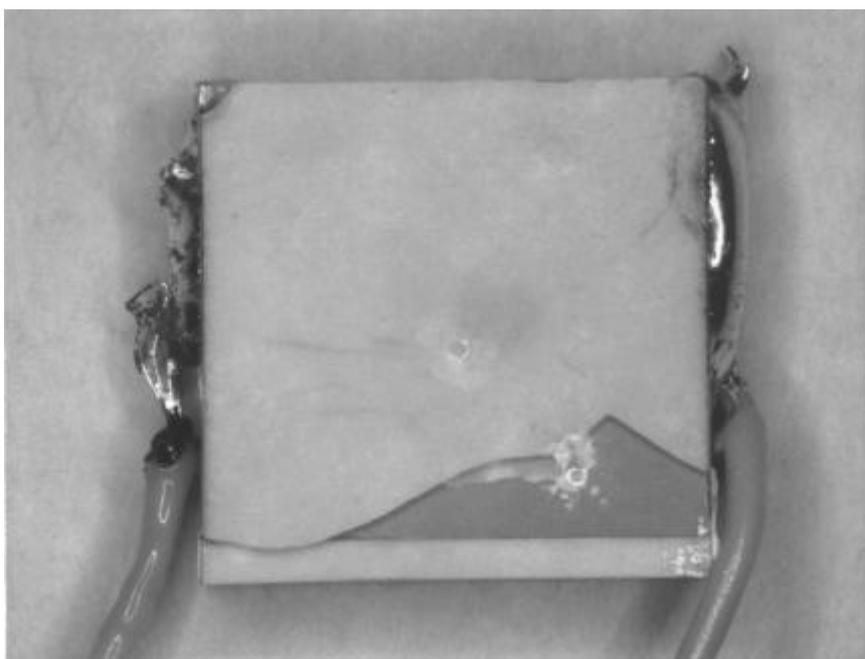


Figure 20: Damage imparted to the multilayer actuator.

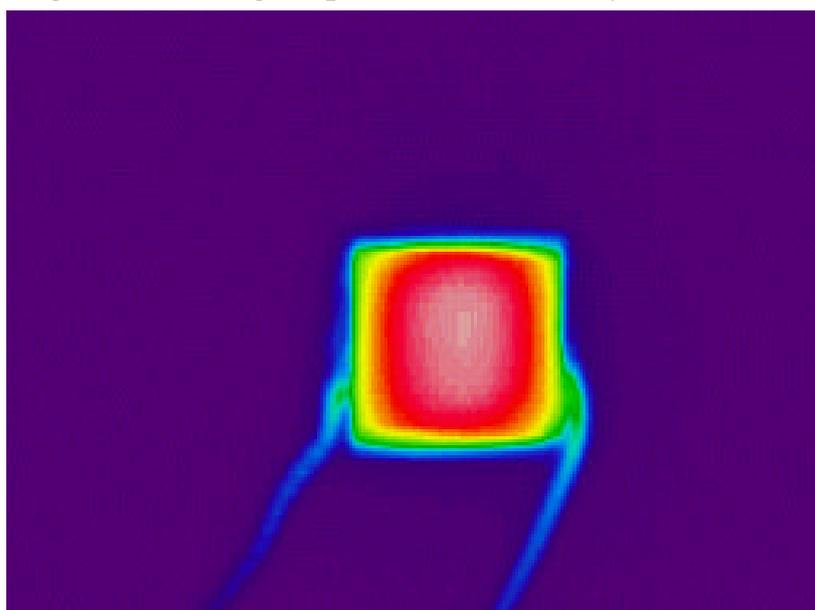


Figure 21: Thermal Image of hard driven actuator of Figure 20.

self heating of the sample and the cooling of the device by the ambient air.

Conclusions

If damage within multilayer actuators is typified by interlaminar or electrode failure then it may be possible that 'hot-spots' may be caused by localised heating caused by shorting electrodes or electrical discharges across the piezoelectric layers. However the thermal signature is dominated by the self heating throughout the thickness of the sample and the likelihood that a small 'hot-spot' in one layer will cause a change in the thermal gradient of the whole sample is small.

In order to detect these defects a much more sensitive detection system will be needed, or perhaps the present methods could be used on much thinner devices, perhaps thick or thin film devices.

References

1. NPL Report CMMT (A) 116, M G Cain, M Stewart, M G Gee, May 1998
2. J. Mandiola and L. Pardo, *Ferroelectrics* **54** (1994) 199.
3. S. Li, C. Y. Huang, A. S. Bhalla and L. E. Cross, *Ferroelectrics Lett.* **16** (1993) 7.
4. C. C. Li, *ferroelectrics* 37 (1981) 623.
5. E. C. Subbarao, M. C. McQuarrie and W. R. Buessem, *J. Appl. Phys.* 28 (1957) 1194.
6. P. Buchman, *Solid-State Electron.* 15 (1972) 142
7. H. Iwasaki and M. Izumi, *Ferroelectrics* 37 (1981) 142.
8. I. J. Mohamad, E.F. Lambson, A. J. Miller and G. A. Saunders, *Phys. Lett* 71A (1979) 115.
9. K. Uchino, T. Hirose and A. M. Varaprasad, *Jpn. J. Appl. Phys.* 26 (supplement 26-2) (1987) 167.
10. W. Pan and H. Cao, *Ferroelectrics* 129 (1992) 119.
11. G. P. Morosova, O. Y. Serdobolskaja, *Ferroelectrics* 75 (1987) 449.
12. T. Hirose, K. Uchino, *Ferroelectrics* 87 (1988) 295; *ibid* 93 (1989) 217.
13. V. Srikanth, E. C. Subbarao, *Acta metall. Mater.* 40 (1992) 1091.
14. V. V. Firsov, A. N. Negovskii, G. G. Pisarenko, V. K. Khaustov, A. M. Aranchii, *Strength of Materials*, 22 (1990) 1455.
15. D. G. Choi and S. K. Choi, *J. Mat. Sci.* 32 (1997) 421.
16. Y. Saito, S. Hori, *Jpn. J. Appl. Phys.* 33 (1994) 5555.
17. Q. Jiang, E. C. Subbarao, L. E. Cross, *Acta metall. Mater.* 42 (1994) 3687.
18. D. C. Lupascu, J. Nuffer, J. Rodel, ISFD-5, PennState, April 6-10. 1998-, *Ferroelectrics* (1998) accepted.
19. H. Aburatami and K Uchino, *Jpn. J. Appl. Phys.*, vol 35, 1996, L516-518
20. CMMT (D)100 M. G. Cain, M. Stewart and M. G. Gee. Actuate SMT Project SMT-CT 95-2029 Mid Term Progress Report, August 1997.
21. T. M. Yonushonis, R. J. Stafford, T. Ahmed, L. D. Favro, R. K. Kuo and R. L. Thomas, *Bulletin Amer. Ceram. Soc.*, 71 [8] (1992) 1191-1202.
22. N K Del Grande, Ph F Durbin, Conference: Twenty-Second Symposium on Quantitative Nondestructive Evaluation, Seattle, 30 July-4 Aug. 1995 Publ. Plenum Publishing Corp., 233 Spring St., New York, NY 10013, USA 1996 15A, 525-531 1995 ISSN: 0-306-45310-X
23. White, G , Mueller, A , Torrington, G, Conference: Twenty-Second Symposium on Quantitative Nondestructive Evaluation, Seattle, 30 July-4 Aug. 1995. Publ. Plenum Publishing Corp., 233 Spring St., New York, NY 10013, USA 1995 REVIEW OF PROGRESS IN QUANTITATIVE NONDESTRUCTIVE EVALUATION 15B, 1961-1967 (1995) ISSN: 0-306-45310-X