

Review of Constitutive Description and Measurement Methods for Piezoelectric, Electrostrictive and Magnetostrictive Materials

R J Von Rensburg*, V Humberstone*, J A Closet, A W Tavernert,
R Stevenst, R D Greenough††, K P O'Connort†, M G Gee
Division of Materials Metrology
National Physical Laboratory
Teddington, Middlesex, TW11 0LW

*The Technology Partnership, Melbown Science Park, Cambridge Road
Melbown, Royston, Herts SG8 6EE

†The University of Leeds, School of Materials, Leeds LS2 9JT

††The University of Hull, Department of Applied Physics, Hull HU6 7RX

ABSTRACT

This report reviews the literature concerned with the constitutive description of piezoelectric, electrostrictive and magnetostrictive materials, and the methods that have been used to characterise their performance. The report is not concerned with a review of the performance or application areas for these materials.

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National Physical Laboratory
Teddington, Middlesex, United Kingdom, TW11 0LW

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Approved on behalf of Chief Executive, NPL,
by Dr M K Hossain, Head, Division of Materials Metrology

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INTRODUCTION

This report gives a review of the literature concerning piezoelectric, electrostrictive and magnetostrictive actuator and sensor materials. The review was performed as part of a project on the development of methods for the high stress characterisation of piezoelectric, electrostrictive and magnetostrictive materials funded by DTI under the MTS Advanced Materials Programme.

The review emphasises two aspects of these materials which are related to their characterisation. These are the extension of their constitutive description beyond the limited linear description which is normally used, and a review of the measurement methods, together with a discussion of their advantages and disadvantages, that have been used for their characterisation.

There are now several introductory textbooks which give some useful information on these materials^{1,2,3,4}. However these are very broad texts, and do not cover measurement and characterisation issues in any great depth. The exception is the compilation by Levinson², which contains a very useful chapter on the strength, fracture and fractography of piezoelectric ceramics. Practical magnetostrictive materials are too new to be mentioned in text books, but useful reviews are given in the papers by Lord and Jiles^{5,6}.

This report has been written as a joint effort by the project partners. Thus section 1 on the Extension of Constitutive Relations for Piezoelectric Materials was written by R W Janse Van Rensburg and Victor Humberstone of the Technology Partnership, Section 2 on the Metrology of Ferroelectric Materials was written by Dr J A Close, Dr A W Taverner and Professor R Stevens of Leeds University, with some significant additions by Dr M G Gee of NPL, and the final section on Magnetostrictive Materials was written by R D Greenough and Kevin O'Connor of Hull University.

SECTION 1: EXTENSION OF CONSTITUTIVE DESCRIPTION FOR PIEZOELECTRIC MATERIALS

The theoretical description of piezoelectric, and more specifically ferroelectric, materials encompasses virtually the entire realm of physics and applied mathematics. The phenomenon of electrically active materials has been known for a long time, beginning with the discovery in the eighteenth century of seignette electricity in what came to be called Rochelle Salt. The fundamental ability of these materials to interconvert electrical, mechanical, thermal and optical energy has resulted in innumerable technological applications. It is therefore not surprising that the amount of literature devoted to the subject is vast and still growing. Some idea of the amount of work concerned explicitly with piezoelectric and ferroelectric substances may be gained from the volumes of Landolt-Börnstein^{7,8} which are dedicated solely to recording their measured properties. This short review will be focused primarily on ferroelectric ceramics and will concentrate only on work which delineates the major developments of the theory.

The physical behaviour of these materials may be summarised in generic terms as the thermo-electromechanical hysteretical dynamics of permanently polarisable polycrystalline materials coupled to external forces and constraints. The study of ferroelectric materials has drawn extensively from each of the fields encapsulated in this descriptive title supplemented by analytical and numerical solutions of the equations governing their behaviour. Naturally each individual field has an extensive literature of its own, but we will consider only a selection relevant mainly to the phenomenology of ferroelectrics. We will concentrate on the latter half of this century, leaving the reader to refer to an account of the early history by Cady⁹.

The earliest studies concentrated on single crystal materials whose symmetries allowed pyroelectric and piezoelectric effects. A very readable presentation of these properties of crystals is given by Nye¹⁰. More sophisticated and highly comprehensive systematic analyses of the crystal classes having piezo- and pyroelectric properties has been carried out, for example, by Aizu¹¹. The lattice distortions which occur when ferroelectrics are polarised, known as the displacive origin of polarisation, have been studied for the better part of this century and an early account can be found in Mason¹². In addition, an explanation of the appearance of a domain structure in a polarised crystal can be given in terms of the reduction in the crystal symmetry at the transition, (see Maugin et al Appendix 1 for a synopsis and references)¹³. A comprehensive discussion of the relation between lattice dynamics and piezoelectricity is given in a review article by Mindlin¹⁴. However, the explanation of piezoelectric behaviour in terms of lattice dynamics remains incomplete to the present, particularly as regards its quantum mechanical origins, see for example Lines and Glass¹⁵. In spite of the complexity of such a fundamental analysis, the lattice dynamical theory of Cochran¹⁶ has been successful in describing the ferroelectric phase transition in terms of a condensing soft phonon, whose frequency goes to zero and which "freezes" at the transition into a lattice distortion which results in a permanent polarisation.

Since a comprehensive fundamental microscopic theory is unavailable and would in any case be extremely difficult to apply, recourse is made to phenomenological descriptions of the material behaviour, as discussed for example in Maugin and Eringen¹⁷ and comprehensively presented by Grindlay¹⁸. These theories are based on the assumption that the material is essentially a dynamical continuum of differentiable fields: matter, electromagnetic, thermal, stress and so on in the usual sense of continuum mechanics. The dynamics of these fields taken separately have been well understood since the last century and are well presented in Landau and Lifshitz^{19,20}, for example. However, their intimate mutual interactions in the interior of a deformable polarisable dielectric body remained largely mysterious until the 1950s when the first rational unified descriptions began to appear, of which the paper by

Toupin²¹ on the elastic dielectric is a recognised classic. On an historical note, it was also about this time that large numbers of ferroelectric materials were discovered, but without at the time having access to a unified theory, see Jona and Shirane²² and the review by Devonshire²³. Later Toupin²⁴ extended his analysis to be fully dynamic. Other seminal works of the period were by Eringen^{25,26} and Truesdell and Toupin²⁷ who published a widely acclaimed article on non-linear field theories in the *Handbuch der Physik*. By 1965 Truesdell and Noll²⁸ could attempt a treatise which would achieve, to use their own word, "permanence" as a comprehensive description of classical fields. Later works of the time served to extend and clarify the formalism, and in particular the work of Tiersten^{29,30} is of note, providing a reference on which the later IEEE standard 176 (1976) on piezoelectric crystals was partly based³¹.

These theories, whether linear or nonlinear, are intended to establish the fundamental dynamical equations of motion, the boundary conditions and the constitutive relations which characterise the material response. In all cases a variational principle is invoked from which the dynamics may be derived. It is also assumed that a thermodynamic potential can be defined in terms of an internal energy and a suitable expression for the free energy, depending on the choice of independent variables. So, for example, Ikeda³² gives a listing of the possibilities, as does Lines and Glass¹⁵. In the linear theories this potential is taken to be a linear function of the independent variables, while in the nonlinear theories it is taken to be a polynomial expansion, a procedure originated by Voigt early in the century, see Cady⁹ or Mason¹² and elaborated upon by many authors. The dependent variables are then derived from the potential by differentiation with respect to their conjugate variables and this gives rise to a set of constitutive relations, see Nye¹⁰ for a straightforward account. Constitutive relations have been intensively studied and a comprehensive analysis is set out in Eringen²⁵. The existence (integrability) of the potential implies a set of reciprocal relations between the material coefficient tensors according to the usual Maxwell relations in thermodynamics. Probably the most advanced form of the variational approach has been developed by Lax and Nelson³³ based on a Lagrangian variation. As a generalisation, the formalism of these theories is of necessity tensorial because of the coordinate invariance requirements and has much in common with the formalism of general relativity. A number of authors such as Grot and Eringen³⁴ and Maugin³⁵ have made this explicit by developing generally covariant formulations.

In general terms the theories mentioned so far are thermodynamically reversible, that is, it is implicitly assumed that changes in the state of the system are carried out in a sequence of equilibrium configurations. However, it is known empirically that ferroelectrics exhibit irreversible behaviours such as hysteresis when subjected to large amplitude periodic disturbances. This hysteresis is commonly ascribed, among other things, to the delays and dissipation which occur when the local polarisation is switched in orientation. We have mentioned that the polarisation occurs in domains in the microcrystals of a ferroelectric ceramic. When an appropriate reversing stimulus is applied these domains switch, not in unity, but by the growth of regularly structured salients in the domain wall from regions of the new polarity into regions of the old. The switching of an individual dipole is intrinsically irreversible and entails, in addition, time delays and losses. A common means, see for example Landau and Lifshitz¹⁹ or Grindlay¹⁸, of describing the hysteresis in the polarisation with respect to the electric field is to adopt a constitutive relation in which the electric field is cubic or higher in the polarisation. Then any given value of the field will generate three roots (or more) for the polarisation, one root (or more) being unstable and two corresponding to the upper and lower branches of the hysteresis loop. However, certain criticisms can be levelled at such a scheme in that, for example, it does not include any time delays and it allows inadmissible states, see Chen and Montgomery³⁹.

Chen and Peercy³⁷ and Chen and Montgomery³⁹ have proposed an alternative approach in which the switching dynamics are made explicit. An evolution equation for the effective

number of aligned dipoles is presented which is added to the other dynamical equations of the system. This effective number of aligned dipoles is akin to an internal variable which determines the strength of the electromechanical coupling coefficients. These authors were able to construct a realistic model of both hysteresis and butterfly loops using only the simplest non-trivial form of their theory. Later Chen^{38,39} extended the theory to be fully three dimensional.

A more sophisticated internal variable approach has been taken by Maugin and co-workers. It is an overt attempt to construct an irreversible, nonlinear theory of hysteresis based on the concepts of virtual power, as extolled by Maugin⁴⁰, the first law of thermodynamics and the second law in the form of the Clausius-Duhem inequality. A comprehensive account of this theory can be found in Bassiouny et al⁴¹⁻⁴³ and in Maugin et al¹³. These authors derive generalised models of hysteresis and show that Chen's theory is included in it. However, it should be pointed out that there are criticisms of the use of the Clausius-Duhem inequality in the sense that the entropy it introduces is not unique, see for example Day⁴⁴.

It is clear that the development of the field is still in progress, but its further advancement will require fundamentally new insights into either its microphysics or into the subtleties of macroscopic irreversible thermodynamics.

SECTION 2: METROLOGY OF FERROELECTRIC MATERIALS

Recent years have seen many new applications arising for piezoelectric, dielectric and electrostrictive devices, which can subject these polycrystalline ceramics to a wide array of stress (both mechanical and electrical), frequency, atmospheric conditions and other constraints. The original limited manufacturers' data relating the performance of their materials is becoming less relevant as the materials are employed under conditions far removed from those under which they were originally characterised. It has therefore become necessary to examine current metrology techniques, to question their applicability and limitations and consequently to develop the techniques whereby characterisation can take place under conditions appertaining to today's demanding applications. Contrary to current practice, the metrology techniques must be such that, in themselves, they do not influence the results obtained. This development will lead to improved product design and performance.

The most common use of the direct piezoelectric effect is for high voltage generation through the application of a compressive stress. Applying a field to the ceramic, the converse effect, enables small movement devices or actuators to be produced. Examples of how this type of behaviour is utilised can be seen in the list of applications below⁴⁵;

Listing of applications for piezoelectric devices.

Impact detonation devices / fuses Gas igniters Active automobile suspension systems Sensors and detectors (transducers) Hydrophones and sonar devices Depth sounders Ultrasonic applications (welding, cleaning, intruder alarms, medical tools) Flaw detectors	Actuators Buzzers Frequency stabilisers Loudspeakers Telecommunications Piezoelectric motors SAW's Medical imaging devices Blood pressure sensors
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Such applications may require the devices to operate at frequencies ranging from quasi-static up to several GHz, produce precision displacement within a few nm and to sustain stresses of hundreds of MPa, although the industrial survey also produced as part of this project shows that much more conservative performance is normally required⁴⁶.

Standards for piezoelectric performance.

Standards for the characterisation and measurement of piezoelectric materials are provided by publications from the Institute of Electrical and Electronic Engineers Inc. (IEEE)³¹, the IRE Standards Committee^{47,48,49} and the International Electrotechnical Commission (IEC)^{50,51}. These standards contain equations based upon the analysis of vibrations developed in piezoelectric materials having simple geometrical shapes. The results from the standards are founded on the assumption of linear piezoelectric behaviour, in which the piezoelectric, dielectric and elastic coefficients (all of which are functions of the state of polarisation) are treated as constants (being termed constants despite the fact we know they are not), independent of magnitude and frequency of the applied mechanical stresses and electric fields⁵².

Unfortunately, mechanical and electrical dissipations (functions of the mechanical strain and electric field respectively) are not introduced into the theoretical treatments and the gross non-linearities germane to ferroelectric materials pertaining to high stress levels are also neglected, as are their hysteretic and ageing characteristics⁵³. The standards involve the dubious practice of combining measurements taken at widely differing frequencies. They do however point out that it is not possible to outline concisely a specific set of conditions under which their definitions and equations apply. It is recommended that the standards will be most applicable to piezoelectric materials with negligible non-linearities and minimal dissipations such as single crystal dielectrics and high coupling factor ceramics (giving rise to the question, how does one know these qualities without measuring and calculating them?).

Unfortunately, this idealistic approach to piezoceramic characterisation is rapidly becoming redundant. Many applications subject ferroelectric devices to large stress (including those thermally induced) and electric bias fields. Compressive pre-stressing in excess of 100 MPa is not uncommon in ultrasonic vibrators and SONAR equipment⁵⁴. Such treatment will induce changes in the piezoelectric, elastic and dielectric behaviour and coefficients will assume an "effective" value. Having said this, it is now common-place for manufacturers to publish graphical data portraying material performance over a limited range of temperature and applied electric field^{55,56,57}. Thus the credibility of the conventional linear equations used to calculate device performance must seriously be brought into question⁵⁸. One should also note that the properties of ceramics are functions of the preparation process and are subject to systematic and statistical fluctuations within a given batch. Proper statistical sampling is necessary to obtain representative "constants" for a given material.

In characterising (and indeed applying, if they are to be used effectively) piezoelectric materials, it is necessary to know and define the electrical and mechanical boundary conditions and make clear **exactly** what has been measured, for instance; the permittivity is dependent on whether the material is free to vibrate or clamped mechanically (i.e., its method of mounting) and the piezoelectric and elastic coefficients are dictated by the electrical circuit into which it is placed (i.e., ranging between open or short circuit conditions)^[15].

Most of the currently available standards are quite old, and it is quite encouraging that there is now new activity in various parts of the world including Europe and Japan to produce new and updated standards for these materials.

Domain effects: Nonlinear characteristics.

As mechanical and electrical stresses vary, ferroelectric materials are subject to deviations from linear behaviour. The limit of linear behaviour is related to the coercive force and varies widely with ceramic composition. Non-linearities are most pronounced under quasi-static stress and short-circuit conditions, therefore such measurements of piezoceramics are subject to considerable variation dependent upon the amplitude of the applied mechanical stress or electric field^{63,64,65}.

It must be pointed out that a number of the formulae exhibited in existing Standards rely on the unwarranted assumption that the Poisson ratio of the ferroelectric materials is constant, whereas in reality, it is also a non-linear function of stress. Whereas the electric field produced by a compressive stress parallel to the poling axis tends to impede domain movement and therefore promotes linear behaviour, a tensile stressing produces a positive bias field and enhances non-linearity. In barium titanate, the generated electric charge density increases more than proportionally to applied stress typically over the range 20 - 100 MPa and then approaches a saturation value equal to the total electric polarisation of the poled ceramic (8 mC cm^{-2}), after which the polarisation state is not recovered upon release of the stress⁶⁶. However, the open circuit field remains a nearly linear function of stress to

far higher levels than does the electric displacement and no permanent loss of polarisation occurs (such measurements must be carried out with a very high impedance data logger). Practical levels of compression e.g., 50 MPa will generate open circuit fields of approximately 15 kV cm^{-1} , but such stresses cause rapid depolarisation in barium titanate compared with hard PZT's which may receive such stresses millions of times without serious degradation. Voltages in excess of 100 kV can be generated by applying (near) destructive compression to poled ferroelectric.

Frequency variation of coefficients becomes most pronounced in ferroelectric materials with relatively high permittivity and compliance loss. The changes in the permittivity, piezoelectric response and compliance may be as high as 5 % per decade of frequency above approximately 1 Hz ⁶⁷.

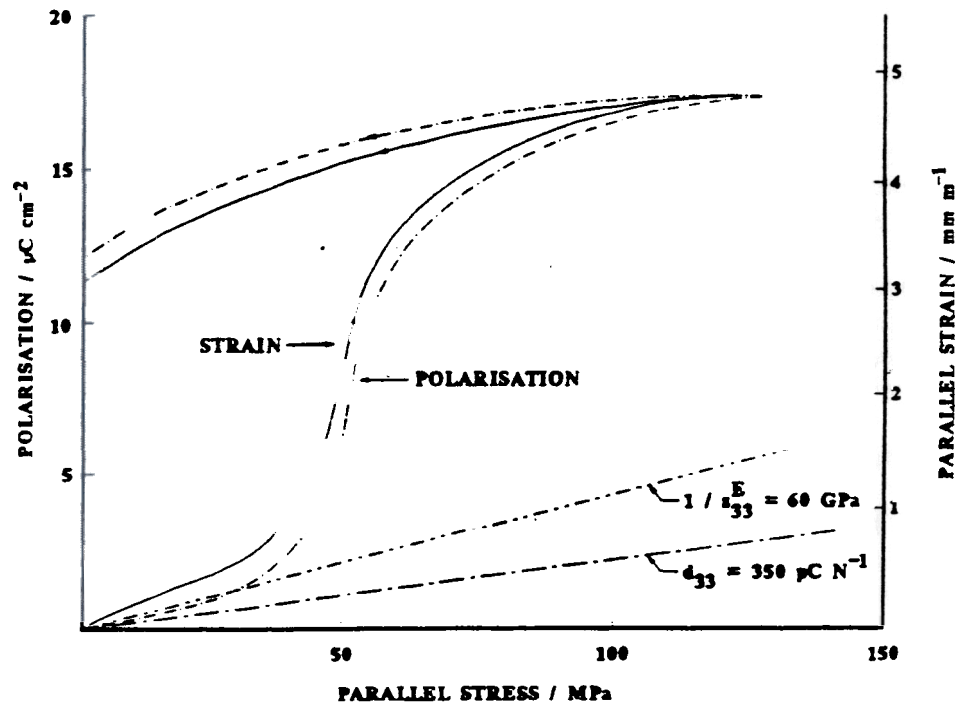
Nonlinear behaviour of piezoelectrics.

Up to a given stress range any charge generated by a piezoelectric material is re-absorbed upon removal of the stress. Below this limit, the stress cycle may be repeated indefinitely without altering the response of the ceramic. The degree of reversibility depends on the rate of application of the stress and the time for which the maximum stress remains applied. The longer a stress is applied, the greater the charge released and the smaller the fraction reabsorbed⁶⁸.

The realisable energy output will depend very much on how the ceramic element is mounted and on the manner in which the stress is applied. If, as is usually the case, the piezoceramic is compressed between two (lower compliance) steel cylinders, lateral clamping and hence barrelling will arise, effectively reducing the value of the piezoelectric coefficient. In the event that the compression cylinder is of higher compliance than the device, for instance if it were of copper, the cylinder end envelopes the end of the device and causes lateral tension, leading to an increased electrical output.

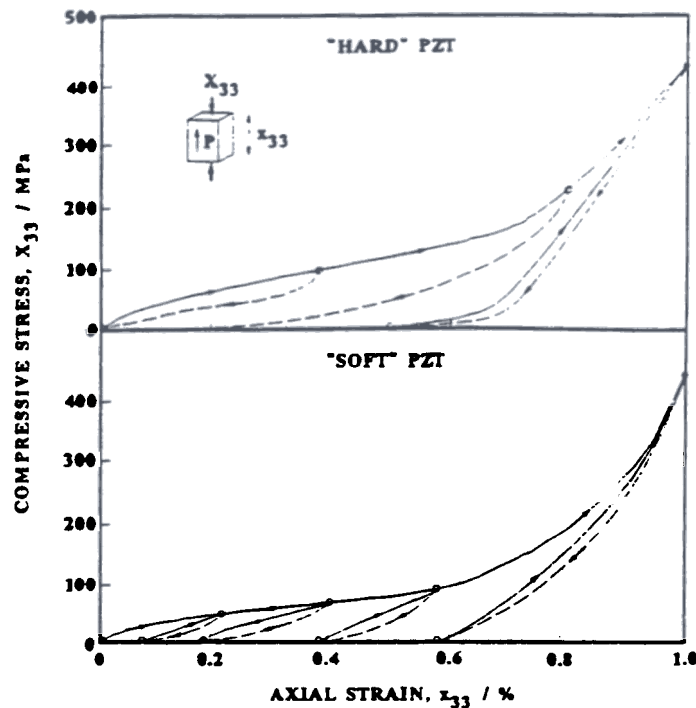
The portion of charge displacement caused by non-linear effects depends to a large extent on the duration of the mechanical stress cycle. If the pulse duration is a few milliseconds or greater, the ferroelectric ceramic still behaves quasi-statically and the charge displacement is non-linear. However, when the stress pulse duration approaches the order of a microsecond, this is short compared with the relaxation times of the ferroelectric domains and the non-linear effects disappear. The charge displacement then corresponds to the linear piezoelectric effect in that it becomes proportional to the applied mechanical stress. This means that higher stresses can be applied for short durations without depolarisation occurring.

It is important to note that non-linear electrical behaviour of the domains, as a function of applied stress, is accompanied by non-linear strain behaviour also as illustrated below. The short circuit output is proportional to the applied mechanical strain, not the stress since it is displacement that produces the direct piezoelectric effect.

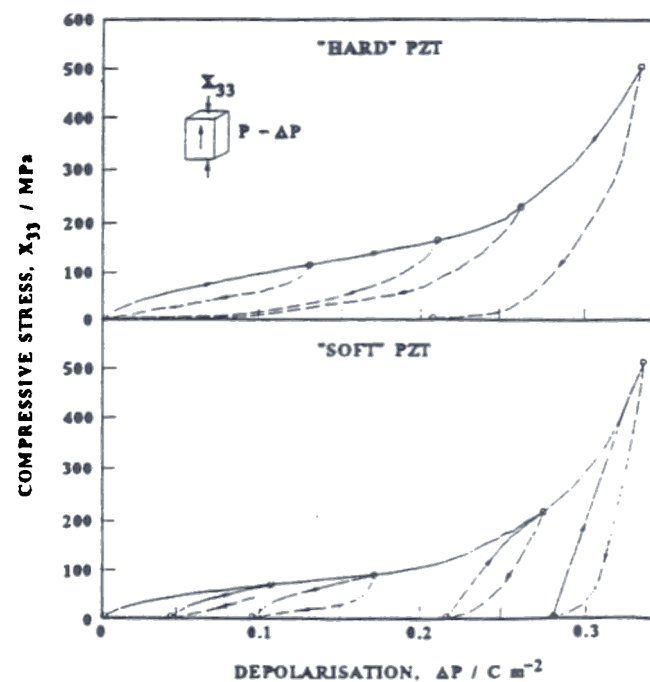


Ferroelectric polarisation and strain as a function of stress^[69]

Most recently, Cao and Evans^{70,71} have embarked on an investigation of the non-linear deformation of ferroelectric ceramics. Both hard and soft PZT materials displayed non-linear deformation behaviour when subject to a compressive stress in excess of ≈ 20 MPa applied parallel to the poling direction. After exposure to a stress above 500 MPa, the materials exhibited a similar permanent deformation of approximately 0.5 % and an 80 % loss of the remanent polarisation state that was induced by the poling process. Complete depolarisation resulted in the material behaving as a linearly elastic body until microcracking commenced prior to mechanical failure. This type of behaviour means that the instantaneous Poisson ratio of the ceramics is a non-linear function of stress, ranging between 0.35 and 0.5. Illustrated below are stress-strain behaviours of the two materials subject to axial stressing. Curves displaying the polarisation change under stress for each material resemble the corresponding stress-strain curves, suggesting that the sources of the non-linear deformation are also responsible for the changes in polarisation. These curves are shown in the figure below. The changes in polarisation and therefore piezoelectric output are, despite being a non-linear function of stress, linearly related to the state of strain in the device.

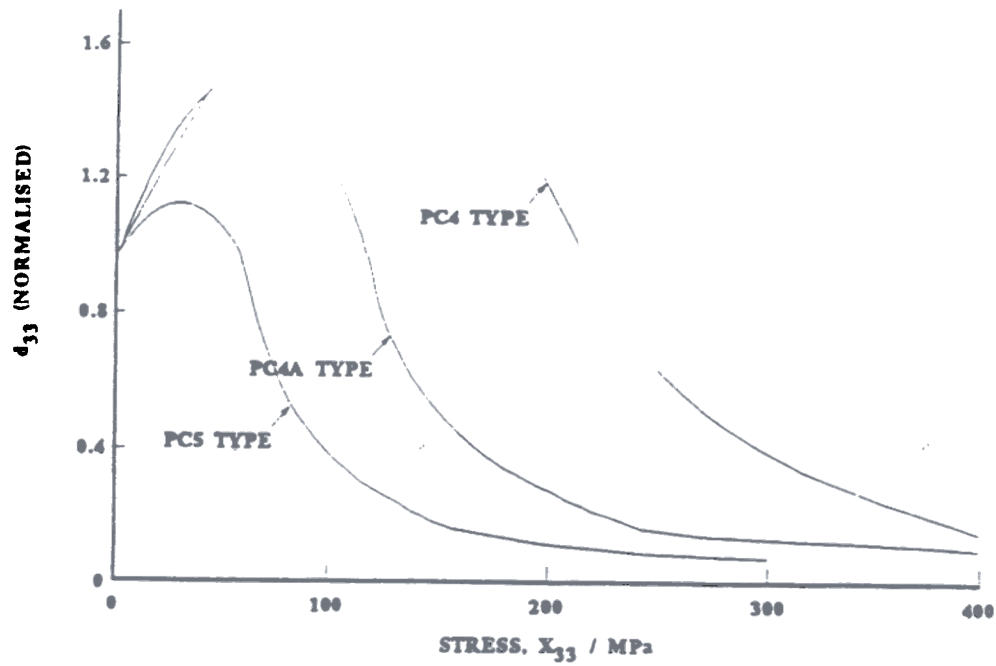


Stress / strain curves for hard and soft PZT ceramics⁷⁰.

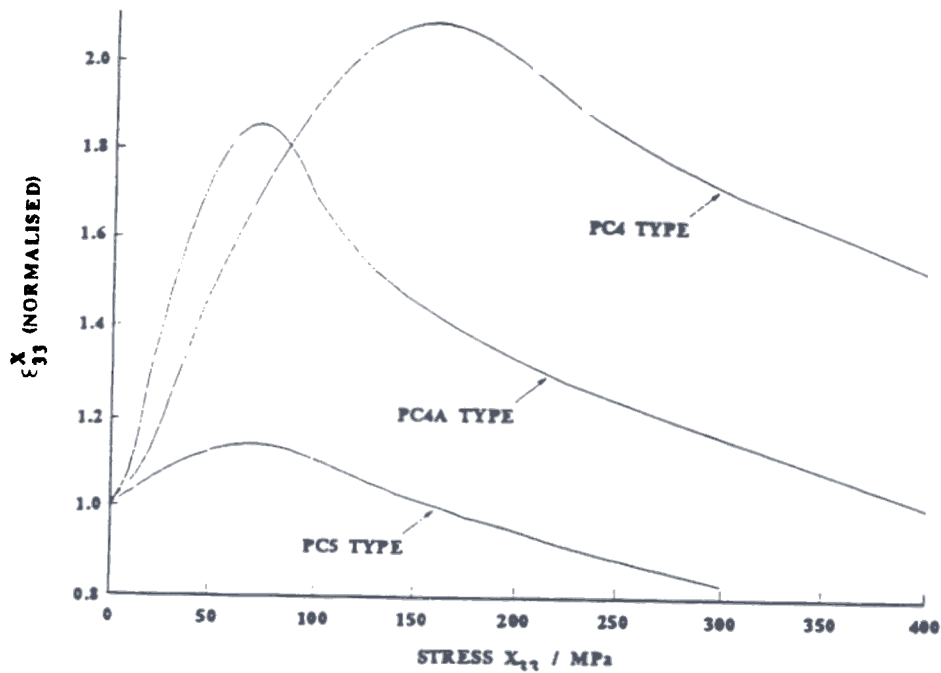


Electrical depolarisation, ΔP as a function of applied stress⁷⁰.

Meekes and Timme⁷¹ carried out extensive investigations on PZT materials akin to the PC4, PC4A and PC5 compositions to show the dependence of g_{33} , d_{33} and ϵ_{33}^x on one dimensional axial stresses (parallel to the poling direction) ranging up to 400 MPa. They observed the onset of a significant, almost exponential drop in the piezo coefficients after stresses reached ≈ 20 MPa with the coefficients of all three materials falling to less than one fifth of their original values at ≈ 300 MPa. The following figures show the normalised charge coefficient as a function of stress for the three PZT compositions and also the normalised values of the relative permittivity under stress⁶⁵.



Effect of quasi-static one dimensional stress on d_{33} [72].



Effect of quasi-static one dimensional stress on relative permittivity.

Basic piezoelectric relationships.

To describe the behaviour of a piezoelectric ceramic with respect to its electrical and elastic properties in terms of the D (dielectric displacement), E (electric field), X (stress) and x (strain), the following equations of state are used:

$$d = \left(\frac{\delta x}{\delta E} \right)_{T,X} = \left(\frac{\delta D}{\delta X} \right)_{T,E} \quad (1)$$

or,

$$g = \left(\frac{\delta x}{\delta D} \right)_{TX} = - \left(\frac{\delta E}{\delta X} \right)_{TD} \quad (2)$$

$$e = - \left(\frac{\delta X}{\delta E} \right)_{Tx} = \left(\frac{\delta D}{\delta x} \right)_{TE} \quad (3)$$

$$-h = \left(\frac{\delta X}{\delta D} \right)_{Tx} = \left(\frac{\delta E}{\delta x} \right)_{TD} \quad (4)$$

Jaffe et al ⁷³ define the relationships for both direct and converse effects as partial derivatives that are evaluated at constant strain (subscript x), constant stress (subscript X), constant dielectric displacement (subscript D) and constant electric field (subscript E). These subscripts represent the boundary conditions "clamped", "unclamped", "open circuit" and "short circuit" respectively. The subscript T denotes constant temperature conditions in all cases (thermodynamic arguments demonstrate that the piezoelectric coefficients for the direct effect are equal to those for the converse effect). Stress changes in a piezoelectric produce a change in the dielectric displacement (the direct effect) and the proportionality constant introduced, d, is the piezoelectric charge coefficient, the subscript E denoting a constant field across the device that is usually zero. By the application of an electric field, a mechanical strain is introduced, this being termed the "converse" effect, the state of constant stress (subscript X) being met by an unconstrained device.

Due to the non-linear characteristics displayed by the ferroelectric ceramics, the basic piezoelectric coefficient relationships are ideally described by the *changes* in the measured parameters:

$$\delta D = d \delta X + \epsilon^X \delta E \quad (5)$$

and;

$$\delta x = s^E \delta X + d \delta E \quad (6)$$

In a poled polycrystalline ceramic, the high symmetry and infinite fold symmetry in a plane normal to the poling direction allows for a reduction of the 18 piezoelectric coefficients to just 3 components which are denoted by the subscripts "31", "33" and "15".

Perhaps the most useful measurement made to characterise a piezoelectric sample is the electromechanical coupling factor, k. On the application of a stress to the device, it is a measure of the fraction of the mechanical energy converted into electrical energy (and vice-versa when the crystal is subject to electrical excitation). This relationship is expressed in terms of k²:

$$k^2 = \frac{\text{mechanical energy converted into electrical energy}}{\text{input mechanical energy}} \quad (7)$$

$$k^2 = \frac{\text{electrical energy converted into mechanical energy}}{\text{input electrical energy}} \quad (8)$$

The value of k^2 and subsequently k , is of course always less than unity since energy conversion is always incomplete since losses necessarily occur.

The coupling coefficients consist of a combination of piezoelectric, dielectric and elastic coefficients and since they are dimensionless, they serve to provide a comparison between differing piezoelectric materials, independent of the specific compliances and permittivity values.

Depolarisation

If the conditions outlined below are not adhered to, depolarisation of the material will occur, i.e., loss of the orientated structure of the electric dipoles, which manifests itself as a reduction or even total loss of piezoelectric properties^[1]: i) The material must be kept at a temperature well below its Curie point. ii) The mechanical stress applied to the material should be within certain specified limits. iii) Excessive alternating electric fields (in both magnitude and frequency) should not be applied to the material. In barium titanate (at room temperature), depolarisation may be detected at 400 V mm⁻¹ and be substantially complete at 800 V mm⁻¹.

Ferroelectric ageing

The properties of a ferroelectric element are subject to time dependence, or "ageing". Following poling of a piezoceramic, the domains may not immediately achieve the lowest possible energy state, due to inhibition of particular domain configurations by lattice imperfections and may even revert to their original directions as a means of stress relief. Depolarising fields are caused by inhomogeneities in polarisation and can also result from the use of semiconducting electrodes¹. The material polarisation as a function of time follows a logarithmic decay and as a result so does the permittivity, piezoelectric coupling factors and elastic compliance.

$$p - p_0 = a \log_{10} \left(\frac{t}{t_0} \right) \quad (9)$$

where $a = a' \log_e 10$ and t_0 is an arbitrary zero time at which point $p = p_0$. "a" varies in sign according to the property; for permittivity it is negative and for Young modulus, positive. The values of a (usually less than 0.05) are most often quoted as a percentage per decade³. Note that the logarithmic law is only an approximation and in the early stages, significant deviations may occur.

Powerful ambient influences that disturb the domain structure cause the ageing rate to deviate rapidly from that predicted until its removal, where it will resume ageing, but at a much advanced stage. Internal heating and high AC and DC fields (showing a time dependence) may partially reverse ageing behaviour. Even heat from the soldering of a device (for instance, a capacitor into a circuit) may temporarily redress the ageing behaviour. Frequently, device testing takes place within a few days of manufacture and so provides values higher than those gleaned at a later date.

Clamping

In a clamped device where free expansion is not permitted, then on application of an electric field, the resulting polarisation is smaller than if it were unclamped. Under clamped conditions, a state of constant (zero) strain exists in the ceramic and when unclamped, the sample is under constant (zero) stress. The relation between the two permittivities existing under these conditions is shown in equation (10) and is related by the previously defined coupling factor, k . In strongly piezoelectric materials, the ratio of free to clamped permittivity may be as great as 2:1.

$$\epsilon_r^x = \epsilon_r^x(1 - k^2)$$

where the coupling factor is given by:

$$k^2 = \frac{s^E d^2}{\epsilon^x}$$

Microstructure

Microstructure plays an important part in the characteristics of a piezoceramic. Aspects to note are grain size, homogeneity of microstructure and amount of porosity present^{74,75}. Crystallite size in a ferroelectric ceramic has a great influence on its dielectric properties. The ferroelectric polarisation becomes locked in if the grains are too small and alignment by an electric field is difficult. This leads to greater dielectric loss and energy dissipation in the ceramic and also to poorer piezoelectric performance. As the grain size increases, the degree of internal stress is reduced, thus removing constraints on domain wall mobility and allowing easier domain movement, resulting in higher values of the piezoelectric coefficients. The limiting grain size is dependent on the material compliance; PZT having a greater compliance than BT, will allow a smaller grain size to be used.

Porosity in a ceramic device may have adverse effects on its performance. The ceramic becomes filled with a multitude of miniature air capacitors leading to possible moisture contamination, dielectric breakdown and reduced electromechanical coupling and piezoelectric coefficients. Inhomogeneities, porosity and flaws lead to local intensification of an applied field, be it electrical or stress, lowering electrical breakdown and mechanical strength. Cracks in the sample surface admit electrode material and provide a spike having a high tip field. Dielectric strength may be dictated by factors such as the electrode configuration, specimen thickness (particularly with respect to flaw size in the device, so sample thickness should be specified when quoting dielectric strength values), temperature, humidity, heat transfer conditions and the operating frequency or time under load of the device. If heat is generated in a dielectric faster than it is dissipated, the resulting temperature rise leads to an increase in conductivity and dielectric loss and eventually, breakdown. Environmental control may be provided by immersing the sample in silicone oil during measurement procedures which are usually carried out at low temperatures. The electrical breakdown of a dielectric device is usually catastrophic and its design must be such as to make it extremely improbable, leading to the necessity for many layers of ground planes. This of course degrades the device's properties and make processing more problematical due to the organic content of the ink. Breakdown strengths in excess of 30 kV mm⁻¹ can be gained in barium titanate materials¹. The most common structural feature limiting breakdown strength is the edge of the electrode. The field at the electrode edge may

be enhanced in a multi-layer structure by the overlap of neighbouring electrodes, where the field may reach a value twice that of the average inside the body of the device.

Ferroelectric ceramics, particularly some of the piezoelectrics are actually structural elements that must withstand quite high stress levels in use. Consequently, knowledge of their mechanical strength is necessary for the production optimum design⁷⁶. Ideally, from a strength standpoint, the ceramics should be used in compression. Mechanical strength and electromechanical behaviour is influenced by internal stresses arising from ferroelastic twinning and anisotropy as a consequence of poling^{77,78}.

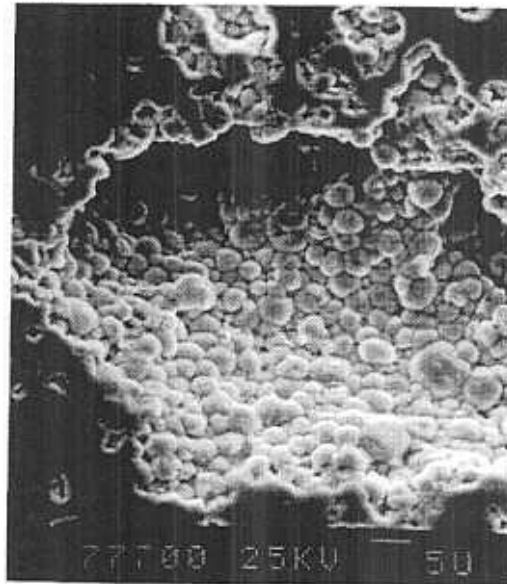
Investigations of fracture mechanics in BT have supported the hypothesis that internal stresses are present in ferroelectric materials. Pohanka et al used a three point bend test to measure the strength of BT^{79,80}. Irrespective of grain size between 2 and 200 μm , the flexural strength at ambient temperature was 60 MPa less than that above the Curie point (typically 135 MPa). The conclusion drawn was that internal stresses, present only below the ferroelectric phase transition, lowered the strength by effectively adding to the applied stress. Pohanka et al. maintain that although stresses in the bulk of each grain are relieved by 90 ° domains, high stress fields are still present at the grain boundaries. Therefore, as grain size decreases, the strength measurements are significantly influenced by the relative increase in grain boundary stresses. Carrying out tests on PZT materials with a grain diameter of 3 μm , Caldwell and Bradt obtained a compressive strength of 650 MPa, compared with 65 MPa under three point bend testing⁸¹. Materials that show the greatest strength and reliability are those that are produced by chemical techniques, where strength improvements of 30 % have been recorded over the conventional mixed oxide route, this being attributed to the highly uniform microstructure⁸².

Manufacturing and testing procedures themselves may to some degree affect the behaviour of a device⁸³. Poling of piezoceramics induces high anisotropic stresses that can lead to crack initiation and subsequent device failure as well as anisotropic elasticity. Testing of dielectric devices may be carried out at up to two or three times the recommended operating levels. Such procedures can induce macroscopic polarisation in the device that affects the behaviour of a capacitor subject to vibration, and there is also the possibility of induced cracking at the interfaces of a multi-layer device.

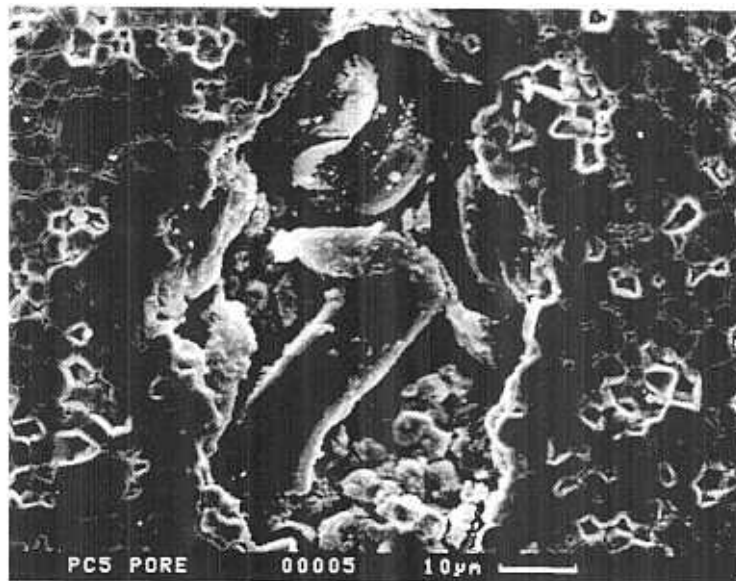
Considering all of the above facts, ultimately, a compromise must be made between the desired piezoelectric and mechanical properties.

Microstructural analysis by scanning electron microscopy

A study of a number of commercial ferroelectric materials⁵⁹ has shown that porosity and contaminants were widespread and provided the initiation sites for fractures under high stress conditions. The occurrence of the pores is a consequence of the pre-sintered powder preparation and firing technique with porosity frequently occurring at what were the interfaces between the spray dried powder particles⁸⁴. Cleanliness of manufacturing route also dictates material behaviour. The two figures below highlight these problems⁵⁹:



Porosity in a PC5 material.

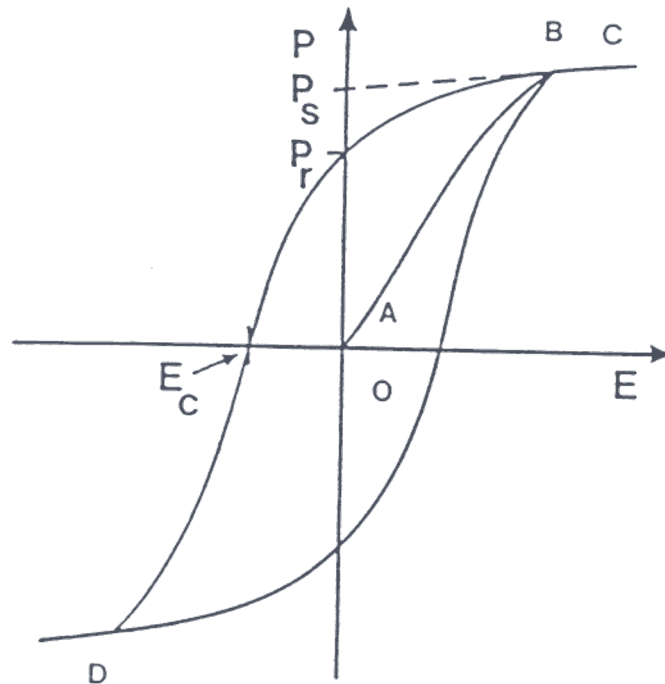


Contaminant present in the PC5 material.

Domain hysteresis and losses.

Intrinsic to the definition of a ferroelectric is the ability to change the direction of the dipole moment by the application of an external electric field. This polarisation reversal is characterised by a hysteresis loop (often seen as proof of ferroelectricity) which arises from the energy needed to reverse the dipoles involving complex geometry and dynamics during each excursion of the field⁶⁶. The area within the loop represents the energy that is dissipated as heat within the sample during the cycle. The hysteresis loop of the ceramic varies

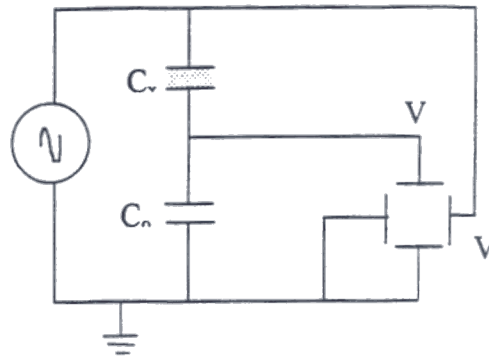
according to the composition, microstructure and internal defects (it has been⁸⁷ suggested that when a field, however small is applied to the sample, it is only a question of time before switching in the crystal occurs). Note that most false reports of ferroelectricity owe their origin to an unwarranted assumption that the resistance of the material was linear and the capacitance was not! Researchers should also be wary of suspiciously horizontal hysteresis loops which may be indicative of saturation of the output signal amplifiers.



Ferroelectric hysteresis loop.

Polarisation- Field hysteresis measurements

Hysteresis measurements on ferroelectric ceramics are conventionally achieved via a Sawyer-Tower type circuit as shown in the figure below.



The 'Sawyer - Tower' circuit

The circuit employs a sample in the form of a plate capacitor, C_x , connected in series with a large integrating capacitor C_o . A sinusoidal voltage is applied across both capacitors, and the voltage appearing across the sample ($V_x \approx V$) is connected across the x-plates of the oscilloscope, whilst the voltage across C_o is connected to the y-plates. Since $Q = C_o V_y$ where Q is the charge on the sample, the trace on the oscilloscope shows the charge on the sample as a function of source voltage. This yields a traditional hysteresis type loop on the oscilloscope.

Several problems exist with this type of approach, however, leading to measurement inaccuracies and poor reproducibility. An obvious problem is deciding the appropriate measurement voltage and whether or not it is really reaching saturation. The onus remains upon the operator to interpret the current-time profile or the *on screen* loop to prevent breakdown and achieve saturation. Another common observation with data generated by this type of conventional *hardware* measurement is that the ends of the loops become rounded near saturation due to linear conductance (resistance) which may include surface and other leakage as well as bulk sample conductance. Finally and probably the most serious problem to the experimental scientist is that the high field ferroelectric saturation region of the loop is inclined - indicative of the residual linear capacitance of the sample.

In order to assess the true ferroelectric hysteretic response of a given sample the linear capacitive and conductive components of the measured signal should be removed or compensated. This is possible via simple hardware means namely variable capacitors and resistors in a duplicate test circuit. Using a differential amplifier, the compensation waveform may then be subtracted from the output of the Sawyer-Tower circuit. Again the process of manually compensating by this method is time-consuming and subject to operator error or variability.

More recently post-data-acquisition processing by computer has been used to carry out the sample current integrations and compensations⁸⁸⁻⁹² with varying degrees of success. These methods, however, fail to tackle the most fundamental problem : accurately determining the saturation conditions. To do this, rather than measuring manually, it is possible to develop *real time* data analysis and control packages capable of continuous compensated loop display. The real time analysis provides for determination of the onset of Ohmic breakdown by

manipulation of the current-time characteristics of the sample as it is measured. The software may then be used to control the source voltage up to an exact point as close to saturation as is possible for a particular sample. Using this method the maximum measuring field is calibrated by a mathematical expression rather than by the operator's eye.

Dielectric behaviour

When under the influence of an alternating field, the charge stored on a dielectric device has both real (in phase) and imaginary (out of phase) components, and it is necessary to account for electrical losses due to energy dissipation by introducing the complex relative permittivity, ϵ^* ⁹³. The complex relative permittivity and its relation to the relative permittivity and losses is described by the relation:

$$\epsilon_r^* = \epsilon_r' - j \epsilon_r'' \quad (12)$$

The term ϵ_r'' is the imaginary component known as the "loss factor" and describes the dielectric loss (average energy stored/energy dissipated per cycle). The losses arising may be represented in terms of "tan δ ", which is defined as the ratio between the imaginary and real parts of the permittivity, where δ is the loss angle. A high loss is undesirable in almost all applications. It lowers the quality factors of resonant circuits and it generates heat, raising the temperature and consequently the behaviour of a device. Invariably, it is assumed that internal dielectric losses are considerably greater than internal mechanical losses.

Ideally, both permittivity and the loss factor should be measured as a function of applied field, frequency and temperature since their behaviour correlates strongly with the freedom of domain wall movement^{62,94}. Dielectric saturation caused by a DC bias voltage may significantly reduce the material permittivity. The low field (unclamped) permittivity and loss are simply determined from the specimen capacitance, C measured at a frequency well below that of resonance - typically 100 or 1 kHz, or a frequency at least 100 times lower than the lowest natural resonant frequency of the element. Poled ferroelectrics show a permittivity that is usually higher than in the unpoled state through contributions from the piezoelectric effect. In ferroelectrics, especially in poled piezoelectric samples, there is a strong dependence of ϵ on frequency and field (high driving fields increase the effective dielectric constant) and typically fields of 1 kV mm⁻¹ are used. The permittivity is found from;

$$\epsilon_{33}^X = \frac{C l}{A} \quad (13)$$

where l and A are the thickness and cross sectional area of the device, under constant stress conditions, denoted by the X superscript. Techniques employing impedance analysers can also give a measure of tan δ , the dielectric dissipation factor, the reciprocal of which is the dielectric quality factor " Q_e ". Measurement of the clamped permittivity proves difficult since effective clamping of a ceramic disc is almost impossible due to its great stiffness⁹⁵. ϵ^x is measured at high frequency (with a thin disc i.e., 1 mm it may be necessary to go to 10 MHz), above all prominent elastic resonances where inertia blocks the piezoelectric effect. Note that typical ferroelectric domain dimensions of 1 mm give elastic resonances into the GHz region where strong dielectric loss is observed. Comparison of the unclamped and clamped dielectric constants would be a convenient way to measure the coupling factors if both permittivities could be measured at the same frequency.

It should be pointed out that device electrodes must be sufficiently thick to avoid the introduction of a series resistance that seriously increases its dissipation factor but must also be thin enough to provide a low inductance, allowing use at high frequency.

Dielectric measurement

The method for measurement of dielectric properties is dependent upon the frequency range of concern. At very low frequencies, usually up to 10 Hz, the current-time curve for charging or discharging the dielectric is used. For medium frequencies of 1 - 10 MHz, the use of a capacitance (Schering) bridge is common, from which the permittivity and loss tangent may be calculated. At very high frequencies exceeding 100 MHz, resonant circuits are employed in which the loss tangent is found from the width of the resonance curve^{88,89}. Commonly, measurements in the low to medium frequency range are accomplished with impedance analysers. These systems consist of two main components, i) a high stability generator, which is used to apply an accurate sinusoidal voltage stimulus to the specimen, and ii) an analyser which resolves the response of the specimen in terms of impedance and phase. Essentially, the analyser operates by applying a sinusoidal voltage across a sample whilst measuring the current transferred across it. Ideally, if the sample were a loss free capacitance, then the current would lead the voltage by a phase angle of 90°. In reality as with any practical capacitor material, there will be a loss within the dielectric, so that the phase angle is less than 90°, differing by the loss angle, δ . The current is resolved into two components; the current in quadrature with the applied voltage, responsible for charging and discharging the capacitor and also the current in phase with the applied voltage, responsible for energy dissipation. These currents are related to the permittivity and loss factor dependent on the nature of the sample impedance. With a knowledge of the specimen geometry and the measurements of the in-phase and quadrature components of current, the capacitance and loss tangent may be found, and it is then a simple matter to derive the relative permittivity of the sample using the elementary equations defining capacitance in terms of specimen surface area, thickness and the permittivity of free space (equation (13)).

Measurements of the dielectric properties under high field conditions are important since there is likely to be a considerable variation in dielectric constant and loss factor with applied field. This can be achieved by means of a specially designed bridge circuit⁹⁶.

The variation in dielectric properties with temperature can be measured by placing the specimen in an appropriate environmental chamber.

Piezoelectric metrology: Resonance techniques

Elastic bodies show numerous resonances; the most pronounced are those where the body can just accommodate one half wavelength of a standing elastic wave. The electrical behaviour of a lightly damped, vibrating mechanical system that is electrically excited by a sinusoidally varying electric field can be to a first approximation represented, in the vicinity of any mechanical resonance, by an equivalent electric circuit^{97,98}. Though the usage of simple idealistic equivalent circuits greatly simplifies the analysis and design of piezoelectric transducers^{99,100}, a number of workers have formulated more accurate equivalent circuits for real systems. However, these are only relevant for the specific situation for which they were developed^{101,102}. A typical electrical circuit comprises a capacitance C_m , a resistance R and an inductance L , shunted by a parallel capacitance C_e . The representation of a piezoelectric vibrator by this circuit is useful only when the circuit parameters are constant and frequency-independent. Generally, the parameters are approximately independent of frequency only for a narrow frequency range close to the resonant frequency and then only if the mode in question is sufficiently isolated from other modes; this being governed by the element geometry. When this is not the case, the equations and measurement methods outlined do not strictly apply.

In a suitably shaped ceramic element, with electrodes perpendicular to and vibrating parallel to the poling direction, specific characteristic frequencies pinpointed as f_r and f_a are the resonant and anti-resonant frequencies respectively. In both cases, the circuit reactance at this point is zero ($X_c=0$). At series resonance ($\approx f_r$), the element has purely ohmic resistance. The element has inductive reactance between series resonance and parallel anti-resonance ($\approx f_a$). At higher frequencies, the element again behaves capacitatively. To a simple approximation, the frequency difference between the series and parallel resonances, Δf , provides a measure of k_{eff} , the effective electromechanical coupling factor of the device i.e., the larger the frequency spread, the greater the coupling. Note that to all intents and purposes, with piezoceramics of high coupling factor and reasonably high mechanical and dielectric quality factors, the frequency differences between the resonant and anti-resonant frequencies and the series and parallel resonances are identical. k_{eff} is given by:

$$k_{eff}^2 = \frac{f_a^2 - f_r^2}{f_a^2} \approx \frac{f_p^2 - f_s^2}{f_p^2} = \frac{C_m}{C_e + C_m} \quad (14)$$

This result is geometry-dependent and k_{eff} is particularly useful in that real materials involve coupled modes due to both piezoelectricity and elasticity effects¹⁰³ and finite lateral dimensions introduce edge effects that may be compensated for with the use of dimensionless shape factors. The aspect ratio of the sample may lead to the interaction of a number of excited vibrational modes, giving rise to a reduced device efficiency. In cases where the intended mode of motion is relatively unrestricted and free from interactions of other modes, specific formulae may be applied to derive the coupling coefficient such as equation 15. A key question is; "in what mode of operation i.e., "33" or "thickness, t", for instance, can the device said to be operating ?". Conflicting ideals are given by differing standardisation bodies^{31,47}. In a device subject to excitation and having its polar axis in the thickness direction, a diameter to thickness ratio of less than 10 is frequently quoted as producing the thickness mode, whereas in cases where the ratio is greater than 5, the operative mode is that of the "33" type, the length extensional mode. The difference is academic since the calculations of both modes are identical. However, since the capacitance of a length extensional element ("33") will be relatively low, care must be taken to avoid stray capacitances.

$$k_{33}^2 = \frac{\pi}{2} \cdot \frac{f_s}{f_p} \tan \frac{\pi}{2} \cdot \frac{\Delta f}{f_p} \quad (15)$$

There exist a number of formulae of this form that are applicable to a particular mode of vibration in the piezoelectric element¹⁹³; k_{31} is simply derived by applying the fundamental f_p and f_s of bars with the thickness along the poling axis and having length to width and length to thickness ratios approximately greater than 3. Though k_t is derived via an identical formula to that for k_{33} as it appears to be the same mode as k_{33} , the value of k_t is smaller than k_{33} due to the constraint imposed by the relatively large lateral dimensions of the disc with respect to its thickness. The measurement is not easily made with accuracy since the thickness resonant frequency is often found split up into several components due to coupling with other bending modes, this difficulty being most pronounced at the anti-resonant frequency. A remedy for the latter problem is measurement of the resonant and anti-resonant frequencies of the 3rd, 5th, 7th or even higher harmonics, which show relatively less splitting and occur at almost exact multiples of the ideal resonances (there is a trivial

[†] For a comprehensive listing of measurement methods and determination of specific coupling coefficients, see references ^{31,47,50}

departure from the true harmonic relation due to the slight variation in acoustic wave velocity with frequency). From these data, the fundamental resonance is readily deduced.

An alternative method of determining the thickness coupling coefficient is from measurement of the dielectric constant well above and well below the thickness resonance, and then applying the approximate equation:

$$\epsilon_3^x = (1 - k_{33}^2) \epsilon_3^X \quad (16)$$

Similarly, the shear electromechanical coefficient k_{15} may be derived by applying ϵ_1^x and ϵ_1^X measured on a bar with a length approximately twice its lateral dimension, poled and electroded along its length.

The planar coupling coefficient, k_p , of a thin disc denotes the coupling between electrical stimulation in the polar (thickness) direction and the simultaneous reaction in the "1" and "2" directions, resulting in a radial vibration (and also therefore, the field produced in the thickness direction by a radially applied stress). k_p is obtained from the fundamental mode f_p and f_s of a thin disc with faces perpendicular to the poling axis⁷³. The applied equations are derived for an infinitely thin disc. For discs of finite thickness, the value of k_p obtained is higher than the true k_p of the disc. Typically, the error arising is less than 1 % for a thickness to diameter ratio < 0.1 . However, the error significantly increases with a reduced aspect ratio, low coupling and high piezoelectric anisotropy. From the measured k_p value, free dielectric constant, ϵ_f and disc density, the piezoelectric constants d_{31} and g_{31} may be calculated.

Compared with single crystal materials, measurements of the dielectric properties of ferroelectrics usually allow one to disregard stray capacitances because of their high permittivities and the relatively high internal mechanical damping (low Q_m) of the materials permits the use of relatively crude specimen holders. However, high electromechanical coupling dictates that complete rather than approximate formulas relating f_p and f_s with coupling factors.

Compliances s_{11}^E , s_{12}^E and s_{15}^D may be determined from the resonance frequencies of bars of specific geometry and electrode configuration, assuming that at fundamental resonance, $c = f\lambda$ and also $c = (\rho s)^{1/2}$ where c is the acoustic wave velocity, $\lambda = 2l$ where l is the governing dimension and ρ is the sample's mass density. A comprehensive outline of the test procedures and apparatus is given by Jaffe et al⁷³.

Measurement of the electromechanical coupling coefficients.

In order to derive the electromechanical coupling coefficient, the resonant and anti-resonant frequencies of the piezoceramic must be determined in agreement with the preceding propositions. In typical measurements, these specific frequencies were found using an impedance analyser, measuring the logarithmic impedance and loss of the DUT as a function of frequency. The DUT was mounted in a binding post fixture with a spring loaded needle point electrode contacting the top surface. This arrangement allows for operation in a (theoretically) unclamped state. Careful analysis of the data is required in order to separate the relevant resonant frequencies from those of other (lateral) vibrational modes and harmonics of the fundamental mode. Additionally, from the resonant frequency, the acoustic wave velocity, c_0 , can be derived using the sample dimensions applicable to that mode of operation. The sample geometry may typically be discs 10 or 20 mm in diameter with thicknesses up to 10 mm, dependent upon the required modes under scrutiny.

Improvements to the present manual procedure for the determination of resonances can be made by computer automation and detection of the points of zero reactance (and gradient of the impedance curve) of a sample subject to a wide frequency scan, typically between 100 kHz and 15 MHz for a 2 mm thick sample. The use of computer algorithms will facilitate the separation of the resonance modes and coupling factors may be determined by the application of the relevant formula and knowledge of the device geometry. It should also be noted many device holders currently in use are not ideally suited to this type of measurement since they do introduce some degree of clamping to the device.

Characterisation by impedance spectroscopy provides information on compositional variation, ceramic texture and defect distribution¹⁰⁴ and analysis of the admittance circles that can be generated from the impedance data yields information of the complex dielectric, elastic and piezoelectric losses¹⁰⁵.

Compliance Measurement.

The elastic compliance of samples in the "33", short circuited mode can be measured by carrying out forced mechanical excitation of a long rod of the sample material, supported on a knife-edge at its midpoint. Rods should have parallel ends and are usually approximately 50 mm in length and 5 mm or less in diameter. The specimen itself is electrically earthed, but close to one end (< 1 mm), an electrode with a bias voltage of approximately 200 volts produces a sinusoidal voltage of a few millivolts. This voltage develops an attractive force between the rod and electrode. The frequency of the periodic voltage may be varied and will, at a given frequency, produce a resonance condition in the excited rod. This state of maximum displacement is sensed by the changing current occurring as a consequence of the changing capacitance by an electrode at the end of the bar distal from the electrode producing the stimuli. From the detected fundamental resonant frequency the vibration wavelength (governed by sample geometry) and the material density; the sample compliance is readily calculated¹⁰⁶.

The elastic compliance of materials can also be measured by the mechanical resonance of bars subjected to single blows, and also by ultrasonic methods¹⁰⁷.

Derivations of further piezoelectric quantities

The parallel resonant frequency is related to the elastic compliance, s_{33}^D by following equation and is measured on open circuit, denoted by the superscript, D:

$$\frac{1}{s_{33}^D} = 4 \rho_0 f_p^2 l^2 \quad (17)$$

where ρ_0 is the mass density of the material and l is the specimen length. It should be noted that the material is more compliant when "shorted" (E is constant) rather than on open circuit, in which case, the relation is given by:

$$s_{33}^E = \frac{s_{33}^D}{1 - k_{33}^2} \quad (18)$$

From the results of two preceding equations, the unclamped permittivity value and k_{33} value obtained from resonance measurements, the piezoelectric charge coefficient may be derived;

$$d_{33} = k_{33} (\epsilon_{33}^X s_{33}^E)^{\frac{1}{2}} \quad (19)$$

and from this result and that of the permittivity, the piezoelectric voltage coefficient is determined via;

$$g_{33} = \frac{d_{33}}{\epsilon_{33}^X} \quad (20)$$

The response to hydro-static pressure introduces the coefficient d_h given by (the 1 and 2 directions are equivalent, therefore $d_{31} = d_{32}$);

$$d_h = 2d_{31} + d_{33} \quad (21)$$

This is measured at a frequency of 10 - 100 Hz¹⁰⁷ by placing the sample in a fluid filled chamber and applying the stress pulse via a piston. Predictably, the hydrostatic voltage coefficient g_h is;

$$g_h = \frac{d_h}{\epsilon^X}$$

An overall measure of the quality of a piezoelectric element may be given, by its figure of merit, M;

$$M = \frac{g_h d_h}{\tan \delta}$$

The mechanical Q factor expresses the ratio of strain in phase with stress to the ratio of strain out of phase with stress as in a vibrating body. The existence of moveable domain walls dominates the level of Q_m which is obtained from the following equation, provided that the minimum impedance $|Z_m|$ at resonance is known:

$$\frac{1}{Q_m} = 2\pi f_s |Z_m| (C_e + C) \frac{f_p^2 - f_s^2}{f_p^2}$$

approximating to;

$$\frac{1}{\omega} \approx 4\pi \Delta f |Z_m| (C_e + C) \quad (25)$$

For this measurement, it is important that the specimen is held only at its nodal point (particularly for high Q materials) since Q_m depends on the device configuration and vibration mode.

The definitive work of Jaffe, Cook and Jaffe (1971)⁷³ contains many derivations and equations for the dielectric, piezoelectric and elastic behaviour of ferroelectric materials that are too voluminous to include here. They describe the experimental prerequisites and data required to describe fully the material behaviour, though in many instances derivations are based again on the assumption of linear behaviour and non-deviatoric material constants.

Piezoelectric charge coefficients

d_{33} coefficients are often assessed under low field conditions at non-resonant frequencies (typically 100 Hz) using a Berlincourt meter¹⁰⁸. Here, the piezoelectric element is lightly clamped between two probes. An electromagnetic driver applies a dynamic force to both the specimen and an internal standard element. Charges generated by both are collected on capacitors and compared by circuitry within the Berlincourt meter, the result appearing as a digital readout. Similarly, appraisal of the d_{31} charge coefficient may be made by mounting the element and subsequently driving and discharging it in a manner which excites the "31" mode.

It is extremely important when measurements of the piezoelectric coefficients are made that the DUT is subject to conditions that are relevant to the piezoelectric equations of state from which they were derived. Bars must be narrow enough and plates wide enough to render unimportant any errors due to the influence of the width-length or thickness-length ratios¹⁰⁹. In general, bars having a thickness-to-length ratio less than 0.1 and plates having a width-to-length ratio less than 0.1 will approximate to an infinitely narrow bar and to an infinitely thin plate respectively. In this way, results from compounded modes of operation may be avoided.

Measuring d_{33} under an applied stress demands that the electric field across the DUT is to be maintained constant (close to zero), though this may not be the case in an application. This condition is ideally imposed by placing a capacitor with at least 10^3 times the capacitance of the specimen across the electroded terminals of the specimen. It is essential to avoid a significant field developing across the sample whilst it is under stress since a build up of charge on the sample faces serves to maintain the state of polarisation of the ferroelectric domains and therefore, by definition, this would impede the generation of current flow from the piezoelectric device. As a result of this, a falsely low value of the piezoelectric charge coefficient would be recorded⁶⁸. The piezoelectric charge coefficients may be measured under static and quasi-static stress conditions with reasonable accuracy, although the precision is inferior to that obtained via dynamic techniques through charge leakage. A useful procedure is shock free lifting of a previously applied weight from the specimen. Again, the charge generated flows into a low loss capacitor, whose capacitance should be several orders of magnitude greater than that of the specimen. When the weight is reapplied, the original voltage reading must be restored if experimental conditions are valid¹¹⁰.

Piezoelectric metrology under impact: Non-linear behaviour.

The non-linear behaviour of ferroelectric materials under rapid, high level stressing has recently been investigated⁵⁹. Samples of commercially produced, poled PZT, with a composition that places them near to the morphotropic phase boundary were subject to longitudinal, compressive impact loading, antiparallel to the poling direction, by a single stress pulse. Six different compositions were tested. Samples were sandwiched in between a Split Hopkinson Pressure Bar instrumented with miniature foil strain gauges, linked to a custom built high frequency strain gauge amplifier. This rig provided tailored, planar stress waves, ranging from 32 to 82 μ s in duration and varying in magnitude between approximately 2 and 40 MPa. The piezoceramics were discharged under short circuit conditions and from these data, values of the piezoelectric charge coefficient, d_{33} were derived and compared with those obtained via the conventional low frequency (100 Hz), low stress (≈ 10 kPa) method.

The electromechanical coupling coefficient, k_t was also derived by correlating the energies produced by the discharging piezoceramic and the stress wave that passed through the sample. These data were then compared with the values calculated from conventional resonance/anti-resonance measurements.

The results obtained from the impact rig gave maximum values for the d_{33} coefficient that were up to twice those provided by the conventional test method¹¹¹. It is apparent that there exists a direct relationship between the impact stress magnitude and the piezoelectric charge coefficient and also the electromechanical coupling coefficient, i.e., the larger the stress, the larger the d_{33} and k_t values, provided that depolarisation of the device had not occurred. It is also apparent that, the shorter the duration of the impact stress wave, the lower the coefficient values, i.e., there exists a frequency dependence. The degree of variation of d_{33} and k_t with stress and impact duration is related to the ferroelectric softness/hardness of the ceramic; the softer materials exhibiting the greatest dependence on these experimental parameters.

As a control and reference exercise, samples of X-cut α quartz were subjected to the same tests as the PZT compositions. The quartz displayed much more stable values of the charge coefficient as a function of impact stress than the PZT materials, but did show a dependence upon the impact stress duration.

It is evident that it is possible within a very wide array, to produce a specific value of the charge coefficient by applying specific impact conditions i.e., stress duration and magnitude, compared with the value suggested by conventional metrology techniques¹¹¹. Examination of the d_{33} versus impact stress curves reveals that the periphery of the plotted lines traces a curve not dissimilar to the cusp of a hysteresis curve of polarisation under an applied field. Since the hysteresis loop of polarisation versus applied field is associated with the characteristic (composition dependent) domain reorientation behaviour, it is suggested that there is a direct link with the domain behaviour and therefore charge flow from the material under stimulation by an applied stress. As the magnitude and time of the stress field applied increases, the degree of domain realignment increases in a manner according to the stability of each type present. The non-linear domain reorientation arises as each individual domain is subject to a different component of the stress field according to its relative crystallographic orientation.

Fatigue behaviour under impact.

Fatigue of the piezoelectric device under repetitive impact stress was also studied by plotting d_{33} as a function of the number of impacts suffered. Samples of the PZT compositions displayed a drop in the values of d_{33} of between approximately 20 and 50 % when subject

to 5000 impacts, each approximately 40 MPa in magnitude and 82 μ s in duration. By comparison the charge coefficient of the quartz had dropped by only 2 % from the original value after 5000 impacts which in the first instance was measured at only 1 % below that gained from the conventional rig. It should be noted that quartz is relatively inert to fluctuations in temperature and ageing with time.

The initially abrupt drop in the d_{33} value is superseded by a flattening off of the decay in d_{33} as the number of impacts progresses. The current drop is a manifestation of the domain switching that has occurred in the ceramic as a means of stress relief, into the more favourable permitted orientations perpendicular to the stressing axis. These results are commensurate with the findings of Bauer et al¹¹² who noted the most marked fatigue behaviour during approximately the first 500 strokes of the impactor onto the PZT materials. The initially rapid decay can be represented as an exponential drop in the charge coefficient under repetitive impact stressing in the poled ferroelectric materials, and good fits to the data are obtained as illustrated by the linearity of the experimental points when plotted on a logarithmic axis. The gradient of the plot, K_i , the rate of change of d_{33} with n_i , may be construed as an ageing coefficient for the material under impact.

$$d_{33} = d_{33}^0 e^{-K_i n_i} \quad (26)$$

A typical commercial piezoelectric igniter that produces approximately 20 kV under an impact of 15 MPa (giving an energy output of 0.05 J m⁻³) is guaranteed a life time of 30,000 strokes during which a 20 % fall in output can be expected¹¹³.

Poled ferroelectric devices are most prone to fatigue behaviour and a loss of piezoelectric properties when i) compressive stress is applied parallel to the poling axis and they are under short circuit conditions since there is no electric field between the electrodes to sustain the domain configurations in their poled position, and ii) under tensile stress parallel to the poling axis as there is a depolarising field produced if open circuit conditions prevail. Also, in view of the frequency dependence of non-linear behaviour, the fatigue behaviour of a ferroelectric device can be expected to be directly related to the duration of loading, be it mechanical or electrical¹¹⁴. Extended application of compression parallel to the poling axis, even at relatively low levels will lead to fatigue of the device through mechanical creep as a means of stress relief⁶¹. The manner of deployment of the piezoelectric device is important, i.e., mode of operation; the electrical and mechanical loading (including its mountings, the constraints it imposes on the device and its direction with respect to the poling axis) put upon it will dictate its fatigue behaviour¹¹⁵. Thus, a poled ferroelectric based device cannot be exposed to different sets of operating conditions and be expected to behave similarly in each instance.

Long term fatigue behaviour

Piezoelectric and electrostrictive actuators and sensors are subject to electrical fatigue as they are cycled in operation. For actuators this is due to the directly applied driving field, and for piezoelectric sensors it is due to the emf generated when they are mechanically cycled.

Cross and coworkers have examined the electrical behaviour of PLZT with composition 7/68/32 under extended cyclic electrical stressing¹¹⁷⁻¹¹⁹. They found several different effects.

Contaminated surfaces caused significant fatigue after 10⁵ switching cycles, but when the cleaning procedures were improved, the samples did not fatigue even after more than 10⁸ switching cycles¹¹⁹. This surface contamination initiated fatigue was explained in terms of the degradation of the interface between the ferroelectric and electrode which led to an inhomogeneous field distribution with eventual microcracking in the ceramic.

In fully dense hot pressed samples no fatigue up to 10^9 cycles was observed.

When porosity was present in the ceramics it was found that the fatigue life was again reduced¹¹⁸. This was felt to be due to pinning of domain wall movement by space charges which accumulated at pores. There was also some more severe damage near the electrode interface.

In large grain samples¹¹⁷, a different failure mechanism occurred due to the development of micro-cracks which evolved into macro-cracks rupturing the sample.

Pan et al have also examined the effect of electric field cycling in ferroelectric ceramic actuators¹²⁰. They found that there was a gradual reduction in the static actuator dimension, an increase in the electric field induced polarisation, and substantial fatigue in electrical field induced polarisation and strain after a number of electric field cycles. This instability varied with the type of ferroelectric ceramic, and the manner with which they are electrically driven. The hysteretic components of the field induced polarisation and strain are the primary cause of the instabilities that were observed.

Several groups have examined the response of ferroelectric materials to extended cyclic mechanical loading. Thus Nishikawa et al¹²¹ and White et al^{122,123} respectively examined the mechanical fatigue life, and growth of cracks under repeated cyclic loading. The growth of subcritical cracks in PZT has also been studied by McHenry and Koepke¹²⁴, who found that the presence of applied fields enhanced crack growth, but that the frequency of application was not important. Crack growth in an electrically stressed multilayer actuator has also been observed using a CCD camera system by Furuta and Uchino¹²⁵.

The effect on piezoelectric performance of mechanical fatigue stressing has also been examined by a number of workers, and a reduction in response has universally been found¹²⁶⁻¹²⁹.

Aburatari and Uchino have also been working on techniques for the non-destructive evaluation of the onset of fatigue degradation¹³⁰. Two promising techniques are the use of acoustic emission monitoring and induced surface potential measurement. It was found that the AE count monitoring was one of the best destruction prediction methods.

Actuators and displacement devices.

Precise positioning by actuator devices can be carried out using electromagnetic, piezoelectric electrostrictive and magnetostrictive materials. Electromagnetic devices suffer from slow response speeds and the ability to exert only relatively low forces compared with the piezoelectric and electrostrictive devices. These can readily produce displacements of $10\text{ }\mu\text{m}$ with an accuracy of 0.1 % in the order of $10\text{ }\mu\text{s}$, even when subject to large opposing stresses in excess of 100 MPa ¹¹⁶.

All materials undergo a small change in dimensions when subjected to an electric field. Unlike piezoelectric behaviour, in electrostrictors, the sign of the deformation (x denoting strain) which occurs with the applied field (E) is independent of the field polarity and proportional to even powers of the field. If ϵ is assumed to be independent of E , although clearly, in the case of ferroelectric and antiferroelectric materials it is a complicated function of E , then applied field and induced strain are related by the electrostrictive coefficient, Q :

$$x = Q \epsilon^2 E^2 \quad (27)$$

giving rise to:

$$x = M E^2 \quad (28)$$

in which M is the electrostriction field coefficient typically having a magnitude of the order of $10^{-15} \text{ m}^2 \text{ V}^{-2}$.

Elastic energy storage may involve high stresses, limited only by mechanical strength, though the relative deformations are small, rarely exceeding 1 part in 1000. A typical piezoelectric bi-morph can be expected to produce a deflection of $1 \mu\text{m}$ when subject to around 100 volts and through lever action this may be multiplied 100 or even 1000 fold¹³¹. The high compliance and low mechanical impedance this causes limit the amount of force it can generate and they are also prone to rapid fatigue¹³². In a sensing application, such a device can be expected to produce of the order of 1 volt. Electrostrictive materials offer some important advantages over piezoelectric ceramics since they do not contain domains and so return to their original dimensions immediately an external field is reduced to zero. By the same reasoning they are less likely to be subject to fatigue or hysteretic behaviour^{116,133,134}. Having said this, the strain produced in the ferroelectric temperature range by electric fields well in excess of the coercive field of a piezoelectric is also independent of the field polarity and thus electrostrictive in nature.

Rapidly usurping the position of bi-morphs and monolithic piezoelectric actuators are macroscopic flextensional transducers that utilise sample and electrode geometry to their advantage¹³⁵, "moonie" devices, "rainbow" monomorphs that utilize material heterogeneity to produce deformation, and also the multi-layer piezoelectric and electrostrictive devices that employ technology akin to that of the multi-layer capacitors. The multi-layer devices afford a maximum displacement (up to $100 \mu\text{m}$) to be obtained from a package occupying the minimum of space and running on a significantly lowered operating voltage (approximately 800 V)^{136,137,138}. Inter-layer spacing ranges between 1 mm and only $20 \mu\text{m}$. However, in these structures, the electric field is non-uniform since the internal electrode area is smaller than the actuator cross sectional area. Consequently, large internal stresses are produced inside the actuator, raising the likelihood of internal mechanical failure that is detectable from the acoustic emission it produces. This behaviour may be alleviated by introducing slits adjacent to the electrodes^{139,140}.

Displacement-field measurement

The measurement of the displacement-field performance of piezoelectric and electrostrictive materials is important to quantify their behaviour in actuator applications. Even before the measurement of displacement is considered, it is important that the sample holding arrangements are properly designed. The use of large clamping forces to hold the sample in position will have an effect on the displacement-field response of the sample. Zhang et al considered this point and found that when silver epoxy was used as a bonding agent, the error introduced would be less than 1% for frequencies less than 20 kHz ¹⁴¹. They also devised a novel vacuum holder.

A number of different systems have been used for the measurement of displacement in the context of these measurements. These include interferometry, strain gauging, capacitance probes, LVDT's dilatometers and other devices.

Interferometers are perhaps the most versatile measurement system as they have a wide dynamic displacement range with resolution down to less than 1 nm , have excellent frequency response, potentially up to several MHz, can have good spatial resolution, and with proper instrumental design can be resistant to the effects of external vibrations¹⁴¹⁻¹⁴⁶.

Strain gauges have been successfully used to measure strain in piezoelectric samples^{59,70,71}, but they suffer from the difficulty that samples are often small, particularly when high field measurements are required, so that the choice and application of appropriate gauging is difficult, and also are very difficult if not impossible to calibrate in a traceable fashion.

Capacitance probes are devices which also have a high dynamic range and high displacement resolution^{147,148}. Their main drawbacks are that they are averaging devices, typically about 10 mm in diameter, and have a relatively low frequency response.

LVDT's have also been used¹⁴⁹ but have a limited displacement resolution and frequency response compared to some other methods, and are therefore perhaps better suited to measurements where large displacements are expected.

A very practical way of measuring the displacement-field performance of these materials at different temperatures is to adopt dilatometers for the measurements¹⁵⁰.

Another method which is proving of considerable interest is to use standoff, non-contact devices such as laser and fibre optic probes¹⁵¹. These devices were often developed for measuring the surface texture of materials, and as well as these advantages accruing from being a no contact measurement method, often have extremely good spatial resolution and frequency response (up to 200 kHz in some cases).

SECTION 3: THE DESCRIPTION OF MAGNETOSTRICTION

This section reviews the state-of-the-art of magnetostriction modelling, acknowledging the fact that the phenomenon is both highly non-linear due to saturation effects, and irreversible due to hysteresis. To this extent a desirable model should be able to predict the shape of minor magnetostrictive hysteresis loops as well as major loops and also account for complex (non-sinusoidal) driving fields where rate dependent effects may be present. Just as importantly, such a model must be based on material parameters which are easily and consistently measured and do not require excessive computation so as to be industrially useful.

As it stands, the vast majority of hysteresis modelling in the literature has been based on magnetic hysteresis, not magnetostrictive hysteresis. All serious attempts at modelling magnetostrictive hysteresis have occurred within the last few years such that it is a relatively young field. However, there are two clear reasons why magnetic hysteresis models may be useful for our purposes:

- (i) Firstly, it has been recognised, physically, that magnetostriction and magnetic hysteresis are intrinsically coupled phenomena¹⁵².
- (ii) Secondly, some of these models are phenomenological in nature so may be readily adapted for magnetostrictive hysteresis.

Notwithstanding, there has been some attempts at modelling magnetostrictive hysteresis in its own right and the generality of these models is steadily increasing. The models fall broadly into three camps.

(i) Jiles, Sablik and co-workers^{152,155,157,161,169,171} have developed a magnetostrictive model as an extension to the Jiles-Atherton model of magnetic hysteresis¹⁷³. This model is based on mean-field equilibrium thermodynamics with additional treatment of domain wall motion. The model is applicable to polycrystalline magnetic materials under uniaxial stress, aligned with the applied magnetic field. In as much, the model is applicable to materials that display sigmoidal shaped hysteresis. The model as it stands presently can, at least qualitatively, yield the behaviour of minor loops for an arbitrary stress (given the constraints above) when supplemented with five experimental parameters. However, it must be stressed that this model is based firmly on equilibrium conditions; it cannot account for rate dependent effects.

(ii) The Preisach hysteresis model is a mathematical construct with a long history in modelling and due to its phenomenological nature, has a wide variety of applications. Subsequently, there is a vast amount of work in the literature about it, attributed to many authors (Mayergoyz is a notable example due to his recent reference book on the subject¹⁶⁴). In the context of this model a magnetic or magnetostrictive material comprises of infinitesimal hysteretic elements. The global properties of the material are given by the interaction of the Preisach elements with both an external field and each other. At present Preisach models of magnetostriction have attempted to describe the effects of applied stress, various non-linearities, rate-dependent effects and both open and close minor hysteresis loops. These models are generally parameterised by characteristic measurements of the major hysteresis loop. The major drawback to the Preisach model is that, unlike the Jiles-Atherton model, it has no firm physical basis. This might make it difficult to derive a general, inter-material model of magnetostriction.

(iii) There have been some attempts to solve magneto-mechanical problems by coupling Maxwell's equations with Newton's second law^{153,160,162,166,170}. The resulting sets of

equations are solved numerically by transforming them to finite difference equations (or similarly using finite analysis), input data being static registrations of the magnetic and mechanical properties. While these approaches can naturally accommodate time-dependent effects (transient behaviour), they do not inherently account for hysteresis. However, Engdahl et al¹⁶⁰ have overcome this by incorporating a stress-dependent Preisach model within their own framework. As such this is one of the more advanced models as it includes both hysteretic and dynamical effects.

Below is the list of references that we consider most relevant and up-to-date for the project.

152 Coupled Magnetoelastic Theory of Magnetic and Magnetostrictive Hysteresis
Sablik, M J, Jiles D C.

IEEE Transactions on Magnetics. Vol 29, No 3, pp 2113-2123, 1993

Comments: An expression for the magnetostriction is derived from minimisation of the internal energy with respect to strains, which is necessary for mechanical equilibrium. Domain wall translation and bowing are accounted for. The model is able to reproduce magnetic hysteresis loops all the way to saturation while providing a framework for describing the effects of stress on hysteresis. In addition, the magnetostrictive features of lift-off and butterfly-shaped hysteresis loops are predicted. The model can accommodate the effects of a stress applied uniaxially to the external magnetic field. It is applicable to polycrystalline magnetic materials under uniaxial stress, aligned with the applied magnetic field.

153 Non-linear Finite Element Modelling of Giant Magnetostriction

IEEE Transactions of Magnetics. Vol 29, No 6, pp 2467-2469, 1993

Benbouzid, M E H, Reyne G, Meunier G.

Comments: A variational formulation is proposed to analyse, by using a finite element method, the giant magnetostriction phenomena. The model is based on non-linear static registrations of the magnetic and mechanical properties of the material. The magneto-mechanical problem is dealt with by coupling Newton's second law with Maxwell's equations. Experimental data are fitted to the model by transforming them into mathematical functions via a surface spline technique. This work takes no account of hysteresis effects.

154 Characterisation of Magnetostrictive Materials with Computer Modelling

Greenough, R D, Reed, I M, Schulze, M P

IEEE Transactions on Magnetics. Vol 29, No 9, pp 2386-2388, 1993

Comments: A plane wave model which enables magnetostriction parameters to be determined is applied to a free standing rod of Terfenol-D. The electrical impedance of a Terfenol rod is simulated so that the magnetostrictive and mechanical properties can be deduced from the impedance characteristics of the whole resonator. By including the frequency dependent losses from eddy currents, the accuracy of the calculated value of k_{33} is enhanced over previous impedance measurement calculations.

155 Numerical Determination of Hysteresis Parameters for the Modelling of Magnetic Properties Using the Theory of Ferromagnetic Hysteresis

Jiles, D C, Thoele, J B, Devine, M K

IEEE Transactions of Magnetics. Vol 28, No 5, pp 27-35, 1992

Comments: The hysteresis parameters in this model are calculated from experimental measurements of the coercivity, remanence, saturation magnetisation, initial anhysteretic susceptibility, initial normal susceptibility and the maximum differential susceptibility. The model deals with magnetic hysteresis by deriving distinct equations for the irreversible and

reversible differential susceptibilities from consideration of domain wall motion. The model shows excellent agreement with experimental data. It is applicable to polycrystalline magnetic materials under uniaxial stress, aligned with the applied magnetic field.

156 Dynamic Generalisation of the Scalar Preisach Model of Hysteresis

Bertotti, G

IEEE Transactions on Magnetics. Vol 28, No 5, pp 2599-2601, 1992

Comments: Presented is a Preisach model of hysteresis in which the rate of change of the flux associated with each elementary loop is proportional to the difference between the external field and the loop threshold fields. This is in contrast to the step-function change in flux associated with the standard Preisach model. This approach should be better at describing dynamic hysteresis phenomena and thus frequency-dependent effects.

157 A Self-Consistent Generalised Model for the Calculation of Minor Loop Excursions in the Theory of Hysteresis

Jiles, D C

IEEE Transactions on Magnetics. Vol 28, No 5, pp 2602-2604, 1992

Comments: The model is based on a simple theory which invokes a constant energy loss per unit change in the magnetisation and thus 'generates' hysteresis from first principles. Specifically, this paper extends an existing model to include a description of hysteresis loops that are not symmetric about zero on the M-H plane (minor loops). This is achieved by invoking an effective volume fraction of active domains determined from the location of the minor loop on the M-H plane. The experimentally observed traits of crossing and partially coincident minor loops emerge naturally from the theory.

158 Numerical Implementation of the Feedback Preisach Model

Mayergoyz, I D, Adly, A A

IEEE Transactions on Magnetics. Vol 28, No 5, pp 2605-2607, 1992

Comments: This model acknowledges the importance of considering feedback in control systems as a result of the demagnetising field phenomenon. Applicable for small feedback factors, the model is non-iterative and thus substantially reduces the complexity of output computations.

159 Energy Considerations in Hysteresis Models

Torre, E D

IEEE Transactions on Magnetics. Vol 28, No 5, pp 2608-2610, 1992

Comments: The aim of this paper is to calculate hysteretic energy loss for open processes (as opposed to closed loops where the energy dissipation is proportional to the area). A Preisach hysteresis model is developed such that there are two distinct output components for the elements; a locally reversible component that can both store and dissipate energy and an entirely dissipative non-reversible component.

160 Application of a Stress-dependent Magnetic Preisach Hysteresis Model on a Simulation Model for Terfenol-D

Kvarnsjo, L, Bergqvist, A

IEEE Transactions on Magnetics. Vol 28, No 5, pp 2623-2625, 1992

Comments: The model is based on static measurements of material characteristics combined with equations for the dynamic mechanical and magnetic behaviour of the material. While incorporating a stress-dependent Preisach hysteresis model, the equations are solved in a finite difference fashion, resolving the transducer in its axial and radial directions. The model

is able to describe the influence of non-linearities, eddy currents and hysteresis on the magnetomechanical transduction process of magnetostrictive materials.

161 Relationship of Magnetostrictive Hysteresis to the Delta-E Effect

Sanlik, M J, Rubin, S W

Journal of Magnetism and Magnetic Materials Vol. 104-107, pp 392-394, 1992

Comments: A new expression for the magnetostriction is derived that is consistent with the conventional formulation for the delta-E effect. A magnetomechanical hysteresis model is employed to investigate magnetostrictive hysteresis under stress and applied magnetic field. The model demonstrates, at least qualitatively, some of the key features observed experimentally.

162 Non-linear 2-D Transient Modelling of Terfenol-D Rods

Kvarnsjo, L, Engdahl, G

IEEE Transactions on Magnetics. Vol 27, No 6, pp 5349-5351, 1991

Comments: This model is based on the static characterisation of the magnetic and mechanical properties of the rods combined with Maxwell's equation and classical mechanical laws. It relies on data obtained from an experimental rig that can produce, independently, both magnetic fields and mechanical stresses of arbitrary shape. A set of finite difference equations are solved by referring to a data base of measured parameters at each time step. Although the model accounts for eddy currents, it does not yet account for hysteresis (Kvarnsjo does this in [9]).

163 Preisach Modelling of Magnetostrictive Hysteresis

Adly, A A, Mayergoyz, I D

Journal of Applied Physics. Vol 69, No 8, pp 5777-5779, 1991

Comments: The authors acknowledge the fact that strain hysteresis has some peculiar properties in comparison with magnetic hysteresis. A Preisach model with two input parameters is developed which the present case represent the variables of magnetic field and stress.

164 Mathematical Models of Hysteresis

Mayergoyz, I D

Springer-Verlag 1991

Comments: A comprehensive overview of Preisach-type models of hysteresis.

165 Preisach Modelling of Hysteresis in Terfenol

Restorff, J B, Savage, A E, Clark A E, Wun-Fogle, M

Journal of Applied Physics. Vol 67, No 9, pp 5016-5018, 1990

Comments: It is stated that Terfenol's hysteresis curves vary markedly from other materials. The model can predict the features of minors loops to an accuracy of about 10-15%. Only the measurement of the first-order reversal curves are required to generate the minor loops - there are no further adjustable parameters. The model is applicable for a fixed longitudinal stress.

166 Non-linear Phenomena in Magnetostrictive Elastic Resonators

El-Nour, A, Alla, A, Maugin, G A

International Journal of Engineering Science. Vol 27, No 12, pp 1613-1619, 1989

Comments: Non-linear vibrations are investigated using the Galerkin method (specifically employed for finite region problems) complemented by a multiple-time scale technique. A set of rotationally invariant continuum equations are introduced. There is exhibited a time-dependent effect called anisochronism due to magnetostrictive couplings at the second order.

167 Dynamic Preisach Models of Hysteresis

Mayergoyz, I D

IEEE Transactions of Magnetics. Vol 24, No 6, pp 2925-2927, 1988

Comments: The numerical implementation of novel Preisach hysteresis models are discussed; they are able to describe dynamic properties of hysteresis non-linearities. Relaxation processes are assumed to be single time-scale processes, although the authors argue that a generalisation to multiple time-scales should be straightforward.

168 Generalised Preisach Model of Hysteresis

Mayergoyz, I D, Friedman, G

IEEE Transactions on Magnetics. Vol 24, No 1, pp 212-217, 1988

Comments: A non-linear Preisach model is developed that has a broader area of applicability than for the classical Preisach model: it relaxes the constraint of congruency (minor loops resulting from oscillatory fields between two extrema are congruent). The non-linear model has the advantage over the classical model that it can fit experimentally measured second-order transition curves.

169 A Model for Hysteresis in Magnetostriction

Sablik, M J, Jiles D C

Journal of Applied Physics. Vol 64, No 10, pp 5402-5404, 1988

Comments: A domain wall pinning model (which includes the effect of an applied stress) is used in conjunction with the Callen and Callen expression for magnetostriction to model magnetic and magnetostrictive hysteresis simultaneously. The model qualitatively explains magnetostriction hysteresis with respect to both H and B.

170 Non-linear Magnetoacoustic Equations

El-Nour, A, Alla, A, Maugin G A

Journal of Acoustic Society of America. Vol 82, No 5, pp 1746-1752, 1987

Comments: Presented in a basic set of rotationally invariant equations of non-linear magnetoacoustics. Applicable in the quasimagnetostatic approximation, the equations are useful to treat non-linear wave-propagation phenomena in centrosymmetric magnetostrictive (such as ferromagnetic polycrystalline materials). The equations are based on the strict use of non-linear continuum mechanics and the electrodynamics of continua with magnetomechanical interactions.

171 Model for the Effect of Tensile and Compressive Stress on Ferromagnetic Hysteresis

Sablik, M J, Kwun, H, Burkhardt, G L

Journal of Applied Physics. Vol 61, No 8, pp 3799-3801, 1987

Comments: A model is presented which qualitatively describes the change in slope and shape of magnetic hysteresis curves with the variation of uniaxial stress. The effect of the

applied stress is treated as an additional contribution to the effective magnetic field in the material and is due to domain vector rotation.

172 Dynamics of Magnetostrictive Ring Transducers

Wilkinson, J P D

Acoustic Society of America. Vol 49, No 5 (part 2), pp 1551-1560, 1970

Comments: Presented is a numerical approach to calculating the dynamic behaviour of magnetostrictive transducers which may have an arbitrary 'shell of revolution' shape. Although the effects of saturation and loss are accounted for, there is no detailed study of hysteresis effects. A set of differential equations are derived, based on Fourier decomposition of the transducers motion.

173 Theory of Ferromagnetic Hysteresis

Jiles, D C, Atherton, D L

Journal of Magnetism and Magnetic Materials. Vol 61, pp 48-60, 1986

Journal of Applied Physics. Vol 55, No 6, pp 2115-2120, 1984

Comments: A hysteresis model based on equilibrium thermodynamics and domain wall motion. Hysteresis is shown to occur as a result of impedances to magnetisation changes such as when domain walls are pinned, while the mutual interactions of magnetic moments are shown to be of secondary importance. Results are presented which show all the features of hysteresis such as the initial magnetisation curve, major and minor hysteresis loops in excellent agreement with (static) experimental results.

174 Galerkin Method for the 3-D Non-linear System of Magneto-thermoelasticity

Novik, O B

Doklady Akademii Nauk Vol 334, pp 100-102, 1994

Comments: Needs translating!

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