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CHARACTERISATION OF PIEZOELECTRIC AND ELECTROSTRICTIVE MATERIALS FOR ACOUSTIC TRANSDUCERS: I. RESONANCE METHODS

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ABSTRACT

Piezoelectric transducers are frequently used as acoustic sensors and projectors as well as in active methods of vibration control. Their proper utilisation requires a good understanding of their non-linear properties and of the dielectric, mechanical and piezoelectric losses in the material. Besides, new computer codes are being developed for the modelling of piezoelectric materials and transducers and these are precise enough to require accurate material constants. The complex impedance of piezoelectric resonators of different geometries can be analysed around their resonances to determine the dielectric, elastic and piezoelectric coefficients as complex constants to take account of all the losses in these materials The impedance curves may be measured as a function of applied dc voltage in order to determine the field dependence of the material constants. By analysing the fundamental and higher resonances, the dispersion in the material constants can be studied and the real and imaginary parts of the constants may be described by frequency dependent polynomials. A new equivalent circuit for the material takes account of all the losses.

INTRODUCTION

Piezoelectric and electrostrictive materials are important constituents of electromechanical sensors, actuators and smart structures. Piezoelectric materials produce a strain, S, under the influence of an external electric field, E, or become electrically polarised under the influence of an external stress, T. The property of piezoelectricity is closely related to the phenomenon of ferroelectricity, which describes the spontaneous polarisation in a crystal that can be changed between two or more distinct directions with respect to the crystal axes through the application of an external electric field. This ability of ferroelectric materials to switch polarisation under an external electric field from a random orientation to a preferred direction is used in a variety of polycrystalline ferroelectric materials (ceramics and polymers) to produce a polycrystalline piezoelectric material with a net preferred polarisation direction This process is described by the term "poling". Prior to poling, individual domains of the ceramic are piezoelectric but the random orientations counteract each other and the net effect is that the macroscopic material shows little or no piezoelectricity. The partial alignment of the domains during poling creates a net spontaneous polarisation in the poling direction and the material shows a C_∞ symmetry around that direction.

Piezoelectricity can be mathematically described by a phenomenological model derived from thermodynamic potentials. The derivations are not unique and the set of equations describing the direct and converse piezoelectric effect depend on the choice of potential and the independent variables used[1,2]. For example, one such set of linear constitutive relations is:

$$
\begin{aligned}
S_p &= s^E_{pq} T_q + d_{pm} E_m \\
D_m &= \varepsilon^T_{mn} E_n + d_{pm} T_p
\end{aligned}
\qquad (1)
$$

where D is the electric displacement, s is the elastic compliance, d is a piezoelectric constant and ε is the dielectric permittivity. The superscripts of the constants designate the independent variable that is held constant when defining the material coefficient and the subscripts define tensor directions which take into account the anisotropic nature of the material. The elements of the tensor form a 9x9 matrix with 1,2,3 designating the orthonormal directions (3 is the poling direction) and 4,5,6 designating the shear directions. For the commonly used polycrystalline piezoelectric ceramic materials with C_∞ symmetry, such as lead zirconate titanate or PZT, there are ten non-zero, independent matrix elements consisting of 5 independent elastic constants, 3 independent piezoelectric constants and 2 independent dielectric constants. For these materials, the reduced matrix form of the above constitutive relationships can now be written as:

$$
\begin{bmatrix} S_1 \\ S_2 \\ S_3 \\ S_4 \\ S_5 \\ S_6 \\ D_1 \\ D_2 \\ D_3 \end{bmatrix}
=
\begin{bmatrix}
s^E_{11} & s^E_{12} & s^E_{13} & 0 & 0 & 0 & 0 & 0 & d_{13} \\
s^E_{12} & s^E_{11} & s^E_{13} & 0 & 0 & 0 & 0 & 0 & d_{13} \\
s^E_{13} & s^E_{13} & s^E_{33} & 0 & 0 & 0 & 0 & 0 & d_{33} \\
0 & 0 & 0 & s^E_{55} & 0 & 0 & 0 & d_{15} & 0 \\
0 & 0 & 0 & 0 & s^E_{55} & 0 & d_{15} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 2(s^E_{11} - s^E_{12}) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & d_{15} & 0 & \varepsilon^T_{11} & 0 & 0 \\
0 & 0 & 0 & d_{15} & 0 & 0 & 0 & \varepsilon^T_{11} & 0 \\
d_{13} & d_{13} & d_{33} & 0 & 0 & 0 & 0 & 0 & \varepsilon^T_{33}
\end{bmatrix}
\begin{bmatrix} T_1 \\ T_2 \\ T_3 \\ T_4 \\ T_5 \\ T_6 \\ E_1 \\ E_2 \\ E_3 \end{bmatrix}
\qquad (2)
$$

While the linear constitutive relations can be written in ways other than shown in (1), there are only 10 independent constants and the IEEE Standard on Piezoelectricity[3] contains the appropriate equations that allow one to convert from one set of equations/matrix to another. Ideally, under small fields and stresses and for materials with low losses within a limited frequency range, these 10 constants contain all the information required to predict the behaviour of the material when a stress, strain or electric field is applied to it In practice most materials display dispersion and non-linearities, have measurable losses and their properties are temperature dependent. Moreover, the properties can have a time dependence due to aging effects.

The most general way to take account of the dielectric, mechanical and piezoelectric losses in a material is to express the 10 material constants as complex coefficients. This paper reviews the experimental methods for determining these complex material constants for any piezoelectric material and the effects of dispersion on these constants, and it discusses the use of an appropriate equivalent circuit for the material to include all types of losses. A second paper[4] reviews the experimental methods to study the non-linear dependence of the material constants on applied signals as well as the response time of the materials

RESONANCE METHODS FOR DETERMINING THE COMPLEX MATERIAL CONSTANTS

The most widely used technique for determining the material constants for piezoelectric materials is the resonance technique as outlined in the IEEE Standard on Piezoelectricity[3]. A piezoelectric sample of specific geometry is excited with an AC signal and an impedance analyser is used to determine the complex impedance and admittance as a function of frequency. Typical spectra are shown in Figure 1 and it can be seen that the spectra contain resonances that result from ultrasonic standing waves in the material. Several particular frequencies may be defined from the spectra: the parallel resonance frequency f_p is the frequency at which the resistance R is a maximum; the sideband frequencies $f_{+1/2}$ and $f_{-1/2}$ correspond to the maximum and the minimum in the reactance X, The series resonance frequency f_s is the frequency at which the conductance G is a maximum; and, the sideband frequencies $f_{+1/2s}$ and $f_{-1/2s}$ correspond to maximum and the minimum of the susceptance B.

The five most common modes used for piezoelectric ceramic analysis are shown in Figure 2 along with the recommended geometrical aspect ratios for samples used to determine each mode. The arrow marked on each sample indicates the poling direction for the piezoelectric ceramics. The aspect ratios ensure that the sample is excited in a mode where the one-dimensional approximation is valid and that coupling between the modes is negligible. In some materials (typically low mechanical Q materials) these aspect ratios may be relaxed whereas in other materials (high Q and high electromechanical coupling) they may need to more stringent.

The impedance equations that govern the various resonance spectra have been derived from the phenomenological theory of piezoelectricity[5,6] for the case of real material constants assuming lossless materials. These equations express the impedance as a function of the appropriate material constants and of the particular frequencies defined above. Holland[7] showed that the losses in a piezoelectric material may be taken into account by representing the material constants as complex coefficients. Sherrit[8] has re-derived the expressions for the

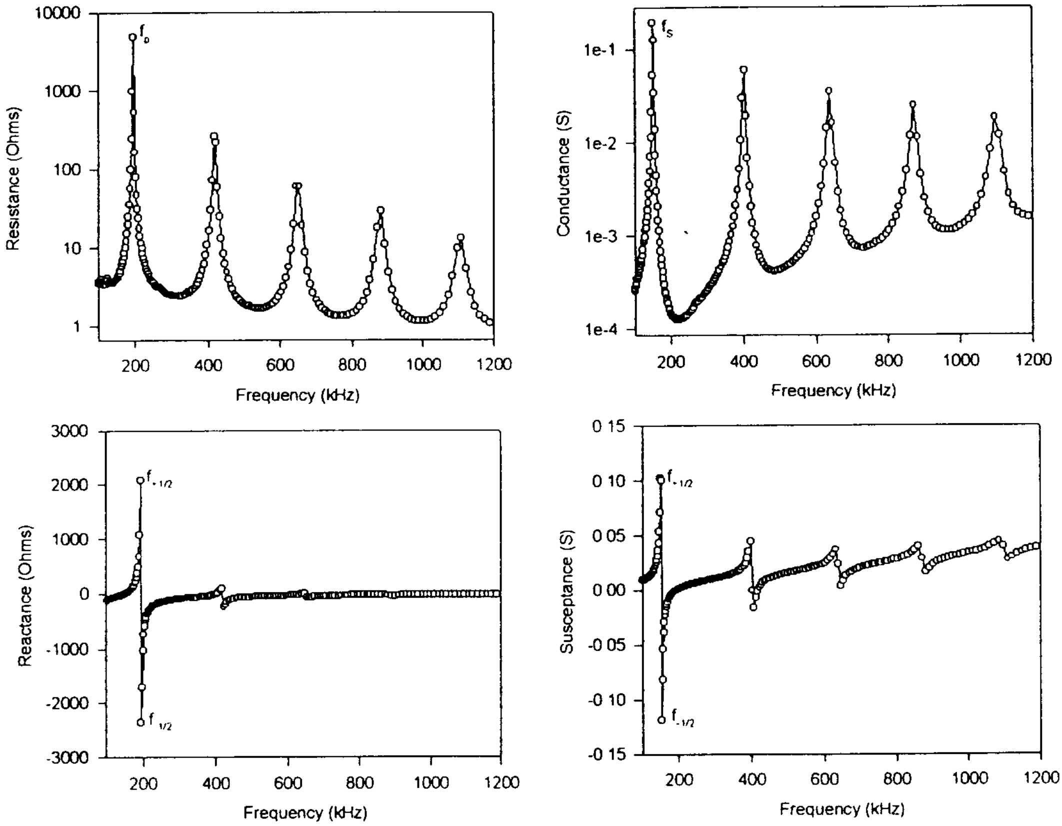

Figure 1. The impedance and admittance spectra for the radial mode of a Motorola 3203 HD PZT disk sample.

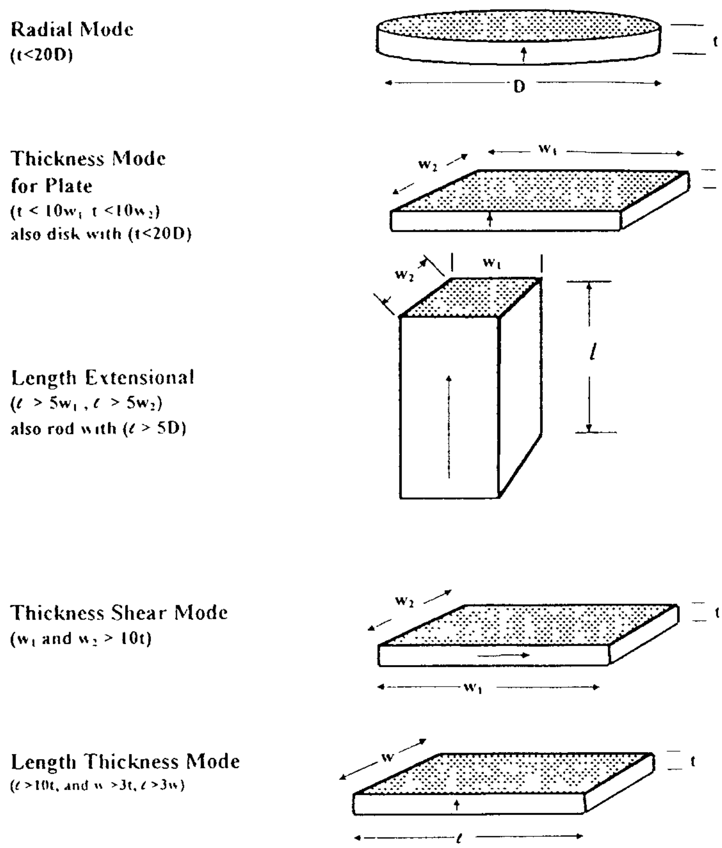

Figure 2. The geometry and poling direction for the five most common modes of piezoelectric resonators for materials characterisation.
Note: The radial mode also has a thickness resonance and the length extensional resonator may also be of the form of a long rod.

impedance for the various resonance geometries using complex material constants so as to include the dielectric, mechanical and piezoelectric losses in the material.

The IEEE Standard on Piezoelectricity uses the impedance equations for lossless resonators and the critical frequencies derived from the equations to determine the real parts of the material constants. Numerous techniques have been proposed to measure the material constants as complex coefficients[9,10,11,12,13,14,15]. Basically, the expression for the impedance is compared with the experimental curve around resonance and the material constants are found so as to obtain the best fit.

As an example, Figure 3 shows the impedance plots for the thickness extensional resonance for Motorola PZT 3203 HD ceramic. For this case the linear piezoelectric equations are

$$T_3 = c_{33}^D S_3 - h_{33} D_3$$

$$E_3 = -h_{33} S_3 + \frac{1}{\varepsilon_{33}^S} D_3 \qquad (3)$$

where h is the piezoelectric constant and c is the elastic stiffness. Considering the material constants to be complex, these equations can be used to derive the expression for the impedance for the thickness extensional resonance:

$$Z = \frac{1}{i\omega A \varepsilon_{33}^S}\left(1 - \frac{k_t^2 \tan\left(\frac{\omega}{4f_p}\right)}{\frac{\omega}{4f_p}}\right), \qquad (4)$$

where the electromechanical coupling constant k_t and the parallel resonance frequency f_p are given by

$$k_t^2 = \frac{\varepsilon_{33}^S h_{33}^2}{c_{33}^D} \qquad 2f_p = \frac{1}{l}\sqrt{\frac{c_{33}^D}{\rho}}$$

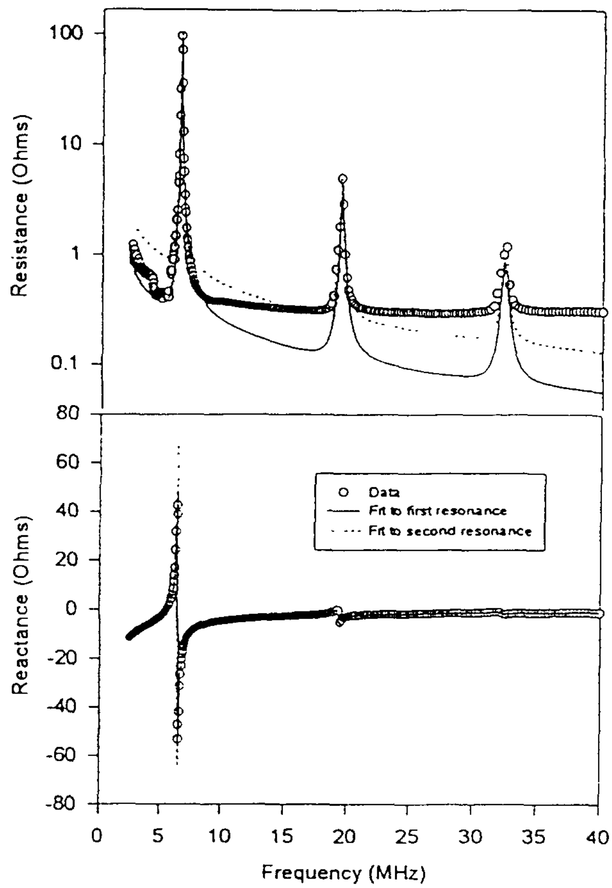

Figure 3. The resistance and reactance data of the thickness extensional resonance and the fit to the first and second resonance peaks for Motorola 3203 HD PZT ceramic. The resistance is plotted on a logarithmic scale

The material constants around the fundamental resonance can be found by fitting expression (4) to the resonance curves shown in Figure 3 This is done by using Smits' method[12] which uses impedance values at three frequencies of which one is chosen near the frequency at which the resistance is a maximum and the other two are chosen to be above and below the resonance. Two of the points plus an initial guess for the elastic constant, using a relationship between the mechanical Q and the resonance bandwidth described by Land et al[16], are used to calculate electromechanical coupling constant and the permittivity. Using the coupling constant and a third point, a new elastic constant is calculated and the process is repeated until convergence. A disadvantage of Smits' technique is that impedance values can vary by several orders of magnitude around resonance and so care must be taken that the measuring instrument is not overloaded and that the impedance values are carefully determined. The values of the material constants will depend to some extent on the points chosen to analyse the spectrum but this dependence can be corrected by repeating the analysis with a different choice of points followed by an averaging of the results Alemany et al[15] have looked at procedures to obtain convergence during iterations. A non-iterative technique to determine the material constants has been presented by Sherrit et al[13]. This technique requires frequency data around resonance and impedance data away from resonance. Although the disadvantage associated with Smits' method is removed, this method is less accurate when dispersion in the material constants is significant. Smits' technique can be used to find the material constants that determine the thickness, thickness shear, length and length thickness modes of resonance. The complex material constants for the radial extensional resonance can be found using a method put forward by Sherrit et al[14]. A commercial software is now available for carrying out the analysis of the resonance curves[17]. By analysing all the different modes of resonance the complete set of material constants can be determined around the resonance considered. An example of such a determination for the Motorola 3203 HD PZT ceramic is shown in Table 1[18].

DISPERSION

The effects of dispersion can be studied by determining the material constants at the fundamental frequency of resonance and at higher resonances. Figure 3 shows that the fit obtained by using the material constants around the fundamental frequency does not agree well with the impedance curve around the second resonance and vice versa. This method has been used to study dispersion in the Motorola 3203 HD ceramic by Sherrit et al[18]. Thus, the material constants can be determined for frequencies required by the applications engineer as long as a reasonable resonance curve can be experimentally determined near the required frequencies. In some cases it is possible to express the real and imaginary parts of the material constant as a polynomial in frequency, as shown by Sherrit et al for piezoelectric polyvinylidene difluoride – tetrafluoroethylene (PVDF-TrFE) copolymer[19]; the applications engineer can use the polynomials to find the material constants at a desired frequency.

EQUIVALENT CIRCUIT

In designing devices it is sometimes useful to have an equivalent electrical circuit to represent the material. Currently the most widely used equivalent circuit to represent a piezoelectric vibrator in the thickness mode is the Van Dyke circuit, which is shown in Figure 4(a). This circuit uses four real circuit parameters, C_0, C_1, L_1 and R_1 to represent the impedance of a free-standing piezoelectric resonator around resonance. However the

Table 1: The reduced matrix of Motorola 3203HD PZT including the electromechanical coupling determined at the fundamental resonance of each mode

Material Constant	Mode	Frequency (kHz)	Value		% Standard Deviation	
			Real	Imag	Real	Imag
$\mathbf{s}_{11}^{E}$ (m^2/N) x10^{-11}	LTE	71.5	1.56	-0.030	0.63	5.2
$\mathbf{s}_{11}^{E}$ (m^2/N) x10^{-11}	RAD	150.9	1.55	-0.032	0.45	2.8
$\mathbf{s}_{11}^{E}$ (m^2/N) x10^{-11}	Average		1.56	-0.031		
$\mathbf{s}_{12}^{E}$ (m^2/N) x10^{-11}	RAD	150.9	-0.420	0.012	3.90	4.9
$\mathbf{s}_{13}^{E}$ (m^2/N) x10^{-11}	Calculated	Smits' formula	-0.821	0.034	N/A	N/A
$\mathbf{s}_{13}^{E}$ (m^2/N) x10^{-11}	Calculated	Matrix inversion	-0.825	0.017	N/A	N/A
$\mathbf{s}_{33}^{E}$ (m^2/N) x10^{-11}	LE	199	1.89	-0.034	1.0	0.78
$\mathbf{s}_{55}^{E}$ (m^2/N) x10^{-11}	TS	2730	3.92	-0.13	2.9	4.3
$\mathbf{s}_{66}^{E}$ (m^2/N) x10^{-11}	Calculated	IEEE formula	3.96	-0.086	N/A	N/A
$\mathbf{c}_{33}^{D}$ (N/m^2) x10^{11}	TE	6390	1.77	0.023	2.0	11
$\mathbf{d}_{13}$ (C/N) x10^{-12}	LTE	71.5	-297	9.7	0.70	7 1
$\mathbf{d}_{13}$ (C/N) x10^{-12}	RAD	150.9	-293	10	0.68	5.8
$\mathbf{d}_{13}$ (C/N) x10^{-12}	Average		-295	9.9		
$\mathbf{d}_{33}$ (C/N) x10^{-12}	LE	199	564	-15	3.1	17
$\mathbf{d}_{15}$ (C/N) x10^{-12}	TS	2730	560	-30	4.6	11
ε_{11}^{T} (F/m) x10^{-8}	TS	2730	2.14	-0.13	0.44	6.8
ε_{33}^{T} (F/m) x10^{-8}	RAD	150.9	3.06	-0.11	1.1	6.5
ε_{33}^{T} (F/m) x10^{-8}	LT	71.5	2.83	-0.061	1.9	9.4
ε_{33}^{T} (F/m) x10^{-8}	Average		2.95	-0 083		
ε_{33}^{S} (F/m) x10^{-8}	TE	6390	1.06	-0.053	2.0	4.2
$\mathbf{k}_{33}$	LE	199	0.763	-0.0029	0.52	45
$\mathbf{k}_{13}$	LTE	71.5	0.447	-0.0054	0.90	16
$\mathbf{k}_{15}$	TS	2730	0.611	-0.0034	3.1	37
$\mathbf{k}_{p}$	RAD	150.9	0.706	-0.0062	0.45	6.1
$\mathbf{k}_{t}$	TE	6390	0.536	-0.0050	0.46	12

impedance, expressed by equation (4), contains six material constants (the real and imaginary parts of the permittivity, the coupling constant and the elastic stiffness) and therefore six parameters are needed to describe the impedance when losses are significant

We have proposed an alternative circuit model based on the lossless resonator model suggested by Butterworth[20] and Cady[21] and shown in Figure 4(b). The model contains three circuit elements C_0, C_1 and L_1 and we modify the model by assuming that each of these circuit elements be considered to be complex. The circuit now has six parameters and some unique features that make it an ideal model for representing the electrical characteristics of an unloaded piezoelectric resonator. We have shown[22] how the values of the complex circuit elements can be calculated from the complex material constants and vice versa and that spectra obtained by using the complex circuit model give very good agreement with corresponding experimental spectra for high Q as well as low Q materials.

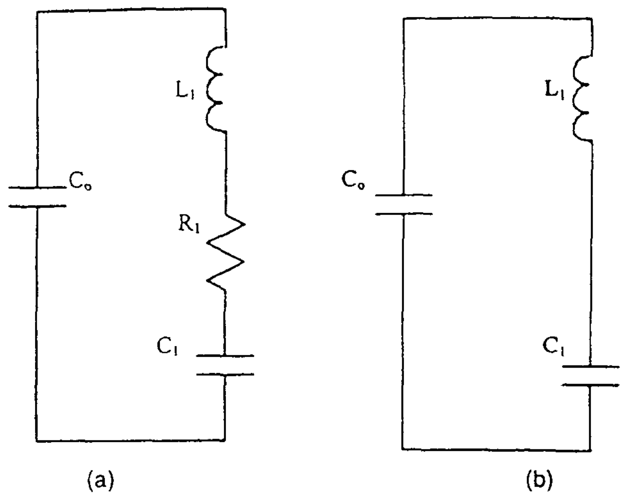

Figure 4. (a) The Van Dyke circuit model; the values of the circuit elements are real. (b) The proposed circuit model; the values of the circuit constants are all complex.

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REFERENCES

[1] A.F. Devonshire, *Phil. Mag. Supp.*, **3**, 85-130 (1952).

[2] W.P. Mason, Physical Acoustics and the Properties of Solids, D. Van Nostrand Co. Inc., (Princeton, New Jersey (19)

[3] IEEE Standard on Piezoelectricity (1987): [ANSI/IEEE Standard 176-1987]

[4] B.K. Mukherjee and S. Sherrit, the next paper in this volume.

[5] D.A. Berlincourt, D.R. Curran and H. Jaffe, Physical Acoustics I Part A: Chapter 3, pp.169-270, Academic Press, Editor: W.P. Mason.

[6] A.H. Meitzler, H.M. O'Bryan and H.F. Tiersten, *IEEE Trans.on Sonics and Ultrasonics*, **SU-20**, 233-239 (1973).

[7] R. Holland, *IEEE Trans. On Sonics and Ultrasonics*, **SU-14**, pp.18-20 (1967).

[8] S Sherrit, Losses, Dispersion and Field Dependence of Piezoelectric Materials, Ph.D Thesis at Queen's University, Kingston, Ontario, Canada (January 1997).

[9] R. Holland and E.P.Eernisse, *IEEE Trans. On Sonics and Ultrasonics*, **SU-16 (4)**, pp.173-181 (1969).

[10] H. Ohigashi, T. Itoh, K. Kimura, T. Nakanishi and M. Suzuki, *Jap. J. Appl. Phys.*, **27**, pp.354-360 (1988).

[11] T. Tsurumi, T. Ichihara, K. Asaga and M. Daimon, *J. Amer. Cer. Soc.*, **73(5)**, pp.1330-1333 (1990)

[12] J.G. Smits, *IEEE Trans. Sonics and Ultrasonics*, **SU-23**, pp.393-402 (1976)

[13] S. Sherrit, H. D. Wiederick and B.K. Mukherjee, *Ferroelectrics*, **134**, pp.111-119 (1992).

[14] S. Sherrit, N. Gauthier, H.D. Wiederick and B.K. Mukherjee, *Ferroelectrics*, **119**, pp.17-32 (1991).

[15] C. Alemany, A.M. Gonsalez, L. Pardo, B. Jimenez, F. Carmona and J. Mendiola, *J. Phys. D: Appl. Phys.*, **28**, pp. 945-956 (1995).

[16] C.E. Land, G.W. Smith and C.R. Westgate, *IEEE Trans. On Sonics and Ultrasonics*, **SU-11**, pp.8-19 (1964).

[17] PRAP (Piezoelectric Resonance Analysis Programme) software available from TASI Technical Software, 174, Montreal Street, Kingston, Ontario K7K 3G4, Canada.

[18] S. Sherrit, H.D. Wiederick and B.K. Mukherjee, Medical Imaging 1997: Ultrasonic Transducer Engineering (SPIE Proceedings Volume 3037), pp.158-169 (1997).

[19] S. Sherrit, J.E. Haysom, H.D. Wiederick, B.K. Mukherjee and M. Sayer, Proc. Of the Tenth Int. Symp. On Applications of Ferroelectrics: ISAF'96, pp.959-962, IEEE, Piscataway, NJ, USA (1996).

[20] S. Butterworth, *Proc. Phys. Soc.*, **27**, pp.410-424 (1915).

[21] W. G. Cady, *Proc. IRE*, **10**, pp.83-114 (1922).

[22] S. Sherrit, H D. Wiederick, B.K. Mukherjee and M. Sayer, *J. Phys. D: Appl. Phys.*, **30**, pp.2354-2363 (1997)