LOSS MECHANISMS IN PIEZOELECTRIC PZT CERAMICS AND SINGLE CRYSTALS

A Thesis in
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by
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ABSTRACT

This thesis aims to clarify the loss mechanisms in piezoelectric ceramics and single crystals in the view of the recent developments in high power piezoelectric devices such as piezoelectric actuators, ultrasonic motors and piezoelectric transformers in commercial applications. In these devices the piezoelectric materials are driven under high voltages and currents. At high power conditions, the materials deviate from their linear constitutive equations due to non-linearity and hysteresis in physical parameters. The non-linearity and hysteresis manifests in terms of loss and consequently, heat generation in piezoelectric materials. Therefore, analysis of the fundamental principles of loss mechanisms is essential in the determination of heat generation. The loss in piezoelectric materials is directly related to the mechanical quality factor which is defined as the inverse of the loss tangent factor. The quality factor is experimentally calculated following IEEE standards using the impedance curves, $Q_m$ as well as near resonance and anti-resonance frequencies and defined as $Q_A$ and $Q_B$. The low power measurement setup in terms of sample holders is reexamined in order to obtain reproducible results. Loss in the 33- and 31- vibration modes is clarified for ‘hard’ and ‘soft’ piezoelectric PZT ceramics. Further loss anisotropy in PMN-PT based single crystals as a function of crystal orientation dependence, vibration mode and doping is investigated. Losses associated with resonance and anti-resonance modes are discussed in the above materials. Significant difference (20-40%) between $Q_A$ and $Q_B$ with $Q$ being higher for anti-resonance mode. This result is verified analytically and dependence of the quality factor on $k_{31}$ (electromechanical coupling coefficient), $\tan\delta'$ (dielectric loss), tan$\phi'$ (mechanical loss) and tan$\theta'$ (piezoelectric loss) is observed. Quality factors ($Q_A$ and $Q_B$) are also calculated at high power near resonance and anti-resonance while maintaining the vibration velocity constant in order to observe the difference at ‘in-service’ conditions. At high power the quality factors drop
by almost half as compared to the low power characterization while maintaining the difference in $Q_A$ and $Q_B$ seen at low power. Based on these results it is recommended to drive piezoelectric devices at anti-resonance mode in order to get better efficiency.
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Chapter 1

Introduction

1.1 Background

1.1.1 Piezoelectricity

In polar dielectric materials, the electric polarization exists in the constituent molecules or atoms, which is randomly oriented. When an electric field is applied, the cations are attracted to the cathode and anions to the anode due to electrostatic interaction. These materials can store more charge due to the dielectric polarization $P$, and the physical quantity of electric charge per unit area is called dielectric displacement $D$. It is related to the electric field by the following expression where the second part is only applicable in a linear dielectric:

$$ D = \varepsilon_0 E + P = \varepsilon \varepsilon_0 E $$

where, $\varepsilon_0$ is the permittivity in vacuum ($8.85 \times 10^{-12}$ F/m),

$\varepsilon$ is the relative permittivity.

Materials in which the centers of positive and negative charges do not coincide even without the application of electric field are said to have spontaneous polarization, $P_s$. When the equilibrium spontaneous polarization of a dielectric material can be reorientated by an electrical field of sufficient strength at temperatures below a characteristic temperature $T_c$, it is called a ferroelectric. Based on the crystallographic symmetry there are 32 different point groups [1] and the structural symmetry in a material affects its dielectric, elastic, piezoelectric, thermal and
optical properties. Further, these 32 point groups can be divided into two groups—(1) 11 centrosymmetric types, which possess center of symmetry and (2) 21 non-centrosymmetric types, which do not have center of symmetry. Out of these 21 non-centrosymmetric group, 20 exhibit piezoelectric effect (except for point group [432]).

When uniform stress is applied in a non-centrosymmetric crystal, there is a movement of positive and negative ions with respect to each other, creating an electric charge at the surface of the material. This phenomenon is the direct piezoelectric effect in which there is a conversion of mechanical energy into electrical energy. In contrast, when an electric field is applied to the crystal, a compressive or tensile strain is produced. This phenomenon is called the converse piezoelectric effect: conversion of electrical energy into mechanical energy. When an alternating electric is applied, mechanical vibration is induced which at appropriate frequency causes mechanical resonance and large resonating strain is created. This phenomenon of strain magnification due to accumulation of input energy is called piezoelectric resonance.

The above direct and converse phenomenon can be represented by the following equations containing stress $X$, strain $x$, electric field $E$ and dielectric displacement $D$:

$$x_i = s^{E}_{ij} X_j + d_{mi} E_m$$  \hspace{1cm} (1.2)

$$D_m = d_{mi} X_i + \varepsilon^X_{mk} \varepsilon_0 E_k$$  \hspace{1cm} (1.3)

where, $i, j = 1, 2, \ldots, 6$; $m, k = 1, 2, 3$ represent the tensor subscripts. Here, $s^E$, $d$ and $\varepsilon^X$ are the compliance at constant electric field, the piezoelectric constant and permittivity at constant elastic stress respectively.
1.1.2 Loss and hysteresis in piezoelectrics

Loss or hysteresis in piezoelectric materials poses a serious problem at off-resonance in positioning actuator applications. Moreover, at resonance, loss generates a significant amount of heat in the piezoelectric materials that causes a serious degradation of device characteristics. The electrical and mechanical losses in ferroelectric ceramics are considered to consist of four portions: (1) domain wall motion, (2) fundamental lattice portion, which should also occur in domain free monocrystals, (3) microstructure portion, which occurs in polycrystals, and (4) conductivity portion [2]. T Ikeda [3] describes the basic formulae for the loss and hysteresis in piezoelectrics. Uchino and Hirose (2001) [4] described the phenomenological theory to deal with losses and developed a method to calculate the intrinsic losses of a piezoelectric ceramic. Loss in piezoelectric materials can be described in terms of electrical and mechanical dissipation factors. These dissipation factors have been included in the piezoelectric material constants as imaginary terms in the complex material constants [Holland et al 1969, 5]. Note that the piezoelectric equations (1.2), (1.3) cannot give the delay-time related loss without taking into account irreversible thermodynamic equations or dissipation functions. This is equivalent to introducing complex parameters $\varepsilon^*$ ($\varepsilon$ is the dielectric constant), $s^*$ ($s$ is the elastic compliance) and $d^*$ ($d$ is the piezoelectric constant) are described in [5] in order to consider the hysteresis losses in dielectric, elastic and piezoelectric coupling energy:
Here, $\delta'$ is the phase delay of the electric displacement to an applied electric field under a constant stress condition, $\phi'$ is the phase delay of the strain to an applied stress under a constant electric field condition and $\theta'$ is the phase delay of the strain under applied electric field or the phase delay of the electric displacement under an applied stress. These phase delays are defined as ‘intensive’ losses. Figure 1.2a-d shows the model hysteresis curves from which we can obtain the above phase delays. In terms of the complex description, the hysteresis loop should be elliptical in shape; however that is not actually observed.

\[ e^{X*} = e^X (1 - j \tan \delta') \]  
\[ s^{E*} = s^E (1 - j \tan \phi') \]  
\[ d^* = d (1 - j \tan \theta') \]
Figure 1.1: (a) $x$ vs $X$ (short-circuit) (b) $D$ vs $E$ (stress free) (c) $x$ vs $E$ (stress free) and (d) $D$ vs $X$ (short-circuit) curves with hysteresis.
The area inside the hysteresis loop corresponds to the energy loss per cycle per unit volume. The stored energies and the losses for pure dielectric and elastic energies are given by:

\[ U_e = \frac{1}{2} \varepsilon^x \varepsilon_0 E_0^2 \]  
\[ w_e = \pi \varepsilon^x \varepsilon_0 E_0^2 \tan \delta' \]  
\[ U_m = \frac{1}{2} s^x X_0^2 \]  
\[ w_m = \pi s^x X_0^2 \tan \phi' \]

The electromechanical hysteresis losses are calculated by measuring the induced strain under an electric field:

\[ U_{em} = \frac{1}{2} (d^2 / s^E) E_0^2 \]  
\[ w_{em} = \pi (d^2 / s^E) E_0^2 (2 \tan \theta' - \tan \phi') \]

Also, electromechanical losses can be obtained by measuring the induced charge under stress:

\[ U_{me} = \frac{1}{2} (d^2 / \varepsilon_0 \varepsilon^x) X_0^2 \]  
\[ w_{me} = \pi (d^2 / \varepsilon_0 \varepsilon^x) X_0^2 (2 \tan \theta' - \tan \phi') \]

Hence from the measurements of above hysteresis loops we can obtain the ‘intensive’ loss parameters i.e. from (D vs. E) and (x vs. X) we can obtain $\tan \delta'$ and $\tan \phi'$; and from the measurements of (x vs. E) or (D vs. X) we can obtain $\tan \theta'$. 
**Losses at piezoelectric resonance**

When an alternating field is applied at an appropriate frequency, there is a mechanical resonance induced in the piezoelectric material. At resonance the mechanical vibration amplitude is amplified by a factor of $Q_m$ (mechanical quality factor) as compared to off-resonance. The expression for admittance in of a piezoceramic can be derived from the expressions of extensional vibration of transverse vibration of piezoelectric plate [1] and is given by

$$Y = j\omega C_d (1 - j\tan \delta) + j\omega C_0 k_{31}^2 \left[1 - j(2\tan \theta' - \tan \phi')\right] \frac{\tan(\omega l/2v^*)}{\omega l/2v^*}$$  \hspace{1cm} \text{(1.15)}$$

where, $\omega$ is the driving frequency,

$C_d$ is the damped capacitance,

$C_0$ is the motional capacitance,

$$C_0 = (wL/t)\varepsilon_0 \varepsilon_x^x, C_d = (1 - k_{31}^2)C_0$$ \hspace{1cm} \text{(1.16)}$$

$\tan \delta$ is the ‘extensive dielectric loss,

$$\tan \delta = \frac{1}{1 - k_{31}^2} \left[\tan \delta' k_{31}^2 (2\tan \theta' - \tan \phi')\right]$$ \hspace{1cm} \text{(1.17)}$$

$k_{31}$ is the electromechanical coupling coefficient,

$v^*$ is the sound velocity in the material,

$$v^* = \frac{1}{\sqrt{\rho \varepsilon_x^x (1 - j\tan \phi')}} = v(1 + j\frac{\tan \phi'}{2})$$ \hspace{1cm} \text{(1.18)}$$

$\rho$ is the density.

Now, the resonance, defined as the maximum admittance point, is derived using the expansion of the term $\tan(\omega L/2v^*)$ around $(\omega L/2v) = \pi/2$ [1]. The maximum motional admittance $Y_{\text{mmax}}$ is obtained as:
\[ Y_{\text{max}}^{\text{max}} = \left( \frac{8}{\pi^2} \right) \omega_0 C_d K_{33}^2 (\tan \phi')^{-1} \]  

(1.19)

Now the mechanical quality factor \( Q_m \) is defined as,

\[ Q_m = \frac{\omega}{2 \Delta \omega} = (\tan \phi')^{-1} \]  

(1.20)

Where, \( \Delta \omega \) is the 3dB bandwidth obtained at \( Y_{\text{max}}/\sqrt{2} \).

Therefore, the mechanical quality factor is inversely proportional to the intensive elastic loss \([1]\).

1.1.3 Piezoelectric materials

The first studies in ceramic piezoelectric materials were done on materials barium titanate (BaTiO₃), PbTiO₃, KNbO₃ for their applications in single crystal and polycrystalline form. However, the discovery of lead zirconate titanate (Pb(ZrₙTi₁₋ₙ)O₃) by Jaffe et al. in 1954 \([6]\) led to widespread usage of piezoelectric polycrystalline materials in various applications. The research into compositional modifications on PZT based ceramics for specific applications led to classification of PZT ceramics into two groups: ‘hard’ and ‘soft’.

‘Hard’ PZT ceramics have acceptor dopants and are characterized by small displacement at off-resonance, low dielectric loss, and high mechanical quality factor \( (Q_m) \). They are therefore used for ultrasonic applications, such as ultrasonic motors. On the contrary, ‘soft’ PZT ceramics are donor doped and present a large strain at off-resonance, high dielectric loss and a low mechanical quality factor. They are generally used in off-resonance devices such as multilayer actuators.
Recently large research effort has been put into Lead Magnesium Niobate-Lead Titanate (PMN-PT) single crystals because of their large electromechanical coupling factors ($k > 0.85$) and piezoelectric constants ($d > 1000 \text{pC/N}$).

### 1.2 Objective

The guidelines to measure the electromechanical constants of piezoelectric materials with low loss have been described in IEEE standards 176-1987. However, without considering the electric and mechanical dissipation, the material properties may be over estimated if IEEE standards are directly used. Smits et al (1976) [7] developed an “iterative-method” based on curve fitting of the theoretical impedance expression to fit three measured data points. Sheritt et al,(1992) [8] developed a method by introducing complex frequencies $f_i$ and $f_p$. This method is limited for materials with low dielectric and piezoelectric loss. Du et al (2000) [9] developed a new method for determination of electromechanical coefficients and extended this approach for single crystals.

Different from the previous studies, this research aims at developing the loss mechanisms in piezoelectric ceramics as well as single crystals based on the experimental method of determining losses from the impedance analysis. Moreover, this study extends over loss dependence on the form factors ($k_{33}$ and $k_{31}$ mode) in ‘hard’ and ‘soft’ piezoelectric ceramics.

In addition, the study is extended to lead magnesium niobate-lead titanate PMN-PT single crystals. Moreover, using the crystal anisotropy it is possible to modify the performance by suitable selection of the orientation. In addition, appropriate composition changes or doping can be used to improve upon the limitations of low thermal or mechanical stability resulting from high loss or low mechanical quality factor $Q_m$. In this
research, we report the loss performance of PMN-PT single crystals as a function of orientation, doping and vibration mode.

1.3 Outline

First the experimental method for impedance measurements to determine the mechanical quality factor is described. In this section, the focus is on obtaining reproducible measurements of low power impedance measurements on piezoelectric materials in $k_{33}$ and $k_{31}$ vibration modes. Based on the experimental techniques developed, the results for piezoelectric ceramics are summarized in chapter 3. The loss for ‘hard’ and ‘soft’ materials and their appropriate drive techniques is described. Next, in chapter 4 the losses in piezoelectric single crystals are described as a function of crystal orientation, doping and vibration mode.

References


Chapter 2

Mechanical Quality Factor

The mechanical quality factor is useful in characterizing resonant systems. Quality factor (Q) compares the time constant for decay of a resonating system’s amplitude to its resonance period. It is generally defined as,

\[
Q = 2\pi \frac{\text{Energy Stored}}{\text{Average Energy Dissipated Per Cycle}}.
\]  

(2.1)

The concept was developed for electrical systems such as filter circuits as well as for mechanical systems such as damped spring-mass systems in order to correlate the sharpness of the respective resonance curve.

2.1 Calculation of Mechanical Quality Factor-\(Q_m\)

As discussed in chapter 1, losses in piezoelectric materials can be divided into three different mechanisms: dielectric, mechanical and piezoelectric losses. These losses are represented in the complex material notation as negative imaginary part [1]. The analytical and experimental method for calculation of these losses was developed by Uchino et al.[2] by extending the intensive and extensive loss model proposed by Ikeda.[3] for the \(k_{31}\) mode. At resonance in piezoelectric materials, the mechanical losses are the most significant one among three losses and it was shown that the mechanical
quality factor $Q_m$, is inversely proportional to the mechanical loss factor, $\tan\phi'$ [4]. Therefore, $Q_m$ value is used as a figure of merit for the losses in piezoelectric materials.

In piezoelectric materials, there is a strong coupling between electrical and mechanical fields so, mechanical, electrical and piezoelectric components of energy losses should be included in the characterization. Generally, in the data sheet available for commercial piezoelectric materials the mechanical quality factor ($Q_m$) and the dielectric dissipation factor ($\tan\delta$) are provided for the loss characteristics. In this case $Q_m$ is calculated near the resonance frequency from a piezoelectric disc in planar vibration mode. [5,6] The elastic, piezoelectric and dielectric constants of a piezoelectric vibrator can be obtained from the resonator measurements by determining the electrical impedance as a function of frequency. Resonance frequency ($f_a$) and anti-resonance frequency ($f_b$), the capacitance and the dissipation factor in the desired frequency range are required to determine the material constants. The methods used for the calculation of $Q_m$ are described below.

### 2.1.1 Equivalent circuit method

In some cases, the material constants are calculated by using the measured parameters from a lumped parameter equivalent circuit as shown in Fig. 2-1. Note that the circuit parameters are assumed constant in the IEEE standards. The method for calculating all the circuit parameters is given in detail in IEEE standard on piezoelectricity.
Once the circuit parameters for the piezoelectric vibrator are determined using an impedance analyzer, the mechanical quality factor can be calculated by using the equation below,

$$Q_m = \frac{1}{2\pi f_r R_1 C_0} \quad (2.2)$$

Where, $f_r$ is the resonance frequency.

2.1.2 Quality factor for $k_{31}$ mode piezoelectric plate

In piezoelectric materials, there exists resonance and anti-resonance as shown in Fig.2-2, which can be understood as electrical short circuit condition and the latter at electrical open circuit condition respectively.
Generally used equations for the determination of the mechanical quality factor of a \( k_{31} \) mode plate sample, as described in the IEEE standards, is obtained by measuring the minimum impedance \( Z_m \) at the fundamental resonance frequency. \( Q_m \) is given by the equation 2.3,

\[
\frac{1}{Q_m} = 2\pi f_a Z_m (C_0 + C_1) \left( \frac{f_b^2 - f_a^2}{f_b^2} \right)
\]

Figure 2-2: Impedance curve of piezoelectric material showing resonance and anti-resonance

where, \( f_a \) is the fundamental resonance frequency, \( f_b \) is the anti-resonance frequency, \( C_0 \) is the shunt capacitance and \( C_1 \) is the series capacitance of the sample.

In the derivation of equation (2.3), it is assumed that \( Q_m \) is exactly same for both resonance and anti-resonance modes, which will be reconsidered in this paper.
2.1.3 Quality factor based on impedance curve

Another way of calculating the quality factor is based on the frequency difference between two frequencies corresponding to the 3 dB below the admittance resonance peak and the impedance anti-resonance peak. Taking narrow frequency range close to the resonance and anti-resonance fig. 2-3, we can define the resonance quality factor ($Q_A$) and the anti-resonance quality factor ($Q_B$) as shown in equation (2.4). In this research, the difference between two quality factors, $Q_A$ and $Q_B$ calculated at resonance and anti-resonance respectively were investigated.

$$Q_A = \frac{f_a}{f_1 - f_2}, \quad Q_B = \frac{f_b}{f_1 - f_2}$$

(2.4)

where, $f_1-f_2$ is the 3 dB bandwidth.

Figure 2-3: (a) $Q_A$ Resonance. (b) $Q_B$ Anti-resonance
2.2 Theoretical analysis

Mezheritsky [8, 9] derived analytical expressions for the quality factors observed at resonance ($Q_A$) and anti resonance ($Q_B$). He describes that there is a difference in these quality factors as opposed IEEE standards which consider $Q_A$ and $Q_B$ as the same. We derived the relationship between $Q_A$ and $Q_B$ on the basis of preceding works by Mezheritsky [8,9]. Consider a piezoelectric rectangular plate in the $k_{31}$ mode. The admittance of this piezoelectric rectangular plate is given by,

\[
Y = j\omega C_d (1 - j \tan \delta) + j\omega C_0 k_{31}^2 \left[1 - j(2 \tan \theta' - \tan \phi') \right] \frac{\tan(\omega l / 2v^*)}{\omega l / 2v^*}
\]  

(2.5)

where,

\[\tan \delta \text{ is the extensive dielectric loss,}\]

\[
\tan \delta = \frac{1}{1 - k_{31}^2} \left[\tan \delta - k_{31}^2 (2 \tan \theta' - \tan \phi') \right]
\]  

(2.6)

\[\tan \theta' \text{ is the intensive piezoelectric loss,}\]

\[\tan \phi' \text{ is intensive mechanical loss,}\]

\[k_{31} \text{ is the electromechanical coupling coefficient and}\]

\[v^* \text{ is the complex sound velocity,}\]

\[v^* = 1 / \sqrt{\rho s_{11}^E (1 - j \tan \phi')} = v(1 + j \tan \phi' / 2)\]

(2.7)
The quality factor at resonance has been derived using the equation (2.5) at its maximum point,[7]

\[ Q_A = \frac{1}{\tan \phi'} \]  

(2.8)

In contrast, the quality factor at anti-resonance \( Q_B \), needs to be derived at the minimum point of equation (2.5). \( Q_B \) is then calculated as,

\[ Q_B = \frac{\omega_b}{\Delta \omega} \]  

(2.9)

where, \( \omega_b \) is the frequency corresponding to \( Y_{\min} \) and \( \Delta \omega \) is the bandwidth \( 2(\omega_b-\omega_1) \), and \( \omega_1 \) is the frequency at which the admittance is \( \sqrt{2Y_{\min}} \). For analysis, we introduce a new parameter \( \Omega = \omega L/2\nu \), normalized frequency:

\[ \frac{\omega l}{2\nu*} = \Omega(1 - j \tan \phi' / 2) \]  

(2.10)

Considering the series expansion of the following equation (2.10),

\[ \tan(\omega l / 2\nu*) = \tan(\Omega - \frac{\Omega \tan \phi'}{2} ) = \tan \Omega - \frac{\Omega \tan \phi'}{2 \cos^2 \Omega} \]  

(2.11)

Using first order approximation for equation (2.11), and substituting equations (2.8) and (2.9) in equation (2.5), we get admittance expression in terms of real and imaginary components. Then define a function \( S \) as a function of \( \Omega \):

\[ S(\Omega) = \left( \frac{L}{2\nu C_0} \right)^2 |Y|^2 \]  

(2.12)

From (2.12), the anti-resonance corresponds to the minimum of \( S(\Omega) \). Now calculate \( S_{\min} \) and obtain the corresponding anti-resonance frequency \( \Omega_b \). The 3 db
bandwidth is obtained by using $S(\Omega')=2S^{\min}$. Then according to the definition, $Q_B$ can be obtained as,

\begin{equation}
Q_B = \frac{\omega_B}{2|\Delta|} \tag{2.13}
\end{equation}

\begin{equation}
Q_B = \frac{(1-k_3^2) + \frac{k_{31}^2}{\cos^2 \Omega_b}}{2(\tan \delta' + \tan \phi' - 2 \tan \theta') + \left[1 + \left(\frac{1}{k_{31}} - k_{31}\right)^2 \Omega_b^2 \right] \tan \phi'} \tag{2.14}
\end{equation}

where, $\Omega_b = \omega l/2\nu$, ($\pi/2<\Omega_b<\pi$). Now considering equation (6) we can get,

\begin{equation}
\frac{1}{Q_B} = \frac{1}{Q_A} + \frac{2}{1 + \left(\frac{1}{k_{31}} - k_{31}\right)^2 \Omega_b^2} (\tan \delta' + \tan \phi' - 2 \tan \theta') \tag{2.15}
\end{equation}

Equation (14) clearly shows the difference between the quality factors at resonance and anti-resonance based upon the parameters, $k_{31}$, $\tan \delta'$, $\tan \phi'$ and $\tan \theta'$. The detailed derivation can be found by Zhuang et.al [12].

2.3 Impedance Analysis

In this section, some guidelines will be introduced regarding the appropriate use of sample holders for impedance analysis. For example, fig. 2-4 shows a sample holder
used for admittance spectrum of \( k_{33} \) mode sample and the corresponding calculated \( Q \) values for the same ‘hard’ piezoelectric sample repeatedly.

Figure 2-4: (a) Sample holder using clamped contacts. (b) Deviation of \( Q \) values over successive readings.

It is seen that the standard deviation is greater than 30% of the mean value. Moreover, measured \( Q \) values are below 600 when the typical \( Q \) value for ‘hard’ piezoelectric sample is greater than 1000 in planar mode. This decrease in quality factor is attributed to the clamping of the sample along the vibration direction. Fig. 2-5 shows the same sample hung freely using thin lead wires.
Figure 2-5: (a) Sample holder contacts through lead wire. (b) Deviation of Q values over successive readings.

In this case, the standard deviation for repeated measurements of the same sample is less than 10% of the mean and the Q values are in tune with the expected for this sample.

The important point in this discussion is that during the impedance analysis, the natural vibration direction of the piezoelectric sample should not be constrained by the contact points of the sample holder. Thus, $k_{33}$ mode bar samples should be freely suspended using thin lead wires and a $k_{31}$ mode plate should be held from the center to allow the two ends to vibrate freely. The recommendations for appropriate sample holders which may be used in impedance analysis of various geometries, is shown in fig. 2-6.
Figure 2-6 Recommendation for sample holder for different geometries.

Using the appropriate sample holders, quality factors were measured for $k_{33}$ and $k_{31}$ mode samples of the same material. The data shown in table 2.1 is from five representative ‘hard’ samples of the $k_{33}$ and $k_{31}$ modes.

<table>
<thead>
<tr>
<th></th>
<th>$Q_A$</th>
<th></th>
<th>$Q_B$</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{33}$</td>
<td>1126±13%</td>
<td>$k_{31}$</td>
<td>1183±12%</td>
<td>Ratio</td>
</tr>
<tr>
<td>$k_{33}$</td>
<td>0.95±6%</td>
<td>$k_{31}$</td>
<td>1868±18%</td>
<td>Ratio</td>
</tr>
<tr>
<td></td>
<td>1868±18%</td>
<td></td>
<td>1986±12%</td>
<td>0.96±25%</td>
</tr>
</tbody>
</table>

All samples were cut to the appropriate dimensions for each vibration mode from the same batch of bulk ceramic and poled at 3kV/mm at 130°C in silicone oil for 30 minutes. The measurements were carried out after 24 hours after poling. It was seen over a range of samples that the ratio of $Q$ values for the $k_{31}$ and $k_{33}$ mode is between 0.9-1.2. This implies that the $Q$ values for $k_{33}$ and $k_{31}$ vibration modes are nearly same and no particular anisotropy in the loss is observed in the two vibration modes.
Chapter 3

Loss in Piezoelectric PZT Ceramics

3.1 Low Power Results

In this section, the quality factor at resonance and anti-resonance are summarized for ‘hard’ and ‘soft’ materials in the $k_{33}$ and $k_{31}$ mode measured using the admittance spectrum from HP4192 impedance analyzer under an oscillation level of 0.5V. Fig. 3.1 summarizes the results for the hard piezoelectric ceramic.
Figure 3-1 Results for ‘hard’ piezoelectric ceramic

Mean $Q_A = 1162$
Mean $Q_B = 1868$

$10 \times 2 \times 2 \text{ mm}^3$

Mean $Q_A = 1183$
Mean $Q_B = 1986$

$17 \times 3 \times 1 \text{ mm}^3$
For both vibration modes, average quality factor of greater than 1000 is observed at the same time it is clear that $Q_B$ is higher than $Q_A$ over all sample range in both $k_{33}$ and $k_{31}$ vibration modes. $Q_B$ is higher than $Q_A$ by 40% for both vibration modes.

Fig. 3.2 summarizes the results for the soft piezoelectric ceramic. Soft piezoelectric material shows quality factor less than 100 and $Q_B$ is higher than $Q_A$ for both vibration modes. However, as opposed to ‘hard’ piezoelectric ceramic, $Q_B$ is higher than $Q_A$ by 35% in $k_{33}$ mode and by 20% in the $k_{31}$ mode.
Figure 3-2 Results for ‘soft’ piezoelectric ceramic.

Mean $Q_A = 100$

Mean $Q_B = 158$

10x2x2 mm$^3$

Mean $Q_A = 75$

Mean $Q_B = 93$

17x3x1 mm$^3$
Figure 3-3 Difference in $Q_A$ and $Q_B$.

Fig. 3.3 shows the difference in $Q_B$ and $Q_A$ in terms of percentage difference. It can be seen that $Q_B$ is 20% to 50% higher than $Q_A$ depending on the material and vibration mode.

### 3.2 High Power Results

In this section, results for high power measurements on $k_{31}$ mode samples are discussed for sake of observing the quality factor at ‘in-service’ conditions for actuators. In this measurement, the vibration velocity is kept constant on the $k_{31}$ mode samples and impedance measurement is carried out using computer control setup [11]. Fig. 9 shows the quality factors as a function of vibration velocity for ‘hard’ piezoelectric ceramic.
As expected, the both $Q_B$ and $Q_A$ drop at higher vibration velocity levels. Moreover, as in the case at low power, $Q_B$ is higher than $Q_A$ at various power levels. Similarly, table 2 shows the quality factor for soft piezoelectric ceramic as a function of vibration velocity.

Table 3.1: Quality factor as a function of vibration velocity for ‘soft’ piezoelectric ceramic

<table>
<thead>
<tr>
<th>Vibration Velocity (mm/sec)</th>
<th>$Q_A$</th>
<th>$Q_B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>78</td>
<td>81</td>
</tr>
<tr>
<td>50</td>
<td>49</td>
<td>63</td>
</tr>
</tbody>
</table>

For soft piezoelectric ceramic the vibration velocity is limited due to large loss and heat generation.
Chapter 4

Loss in PMN-PT Single Crystals

4.1 Background

The figures of merit for sonar transducers are high bandwidth, high power efficiency and high energy density. These device performance parameters are a function of electromechanical coupling factor, piezoelectric coefficient and drive level limits of the material (i.e., maximum vibration velocity). High electromechanical coupling factors and high piezoelectric constants have been reported in some relaxor ferroelectric single crystals. For example, in Pb(Zn$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO$_3$ (PZN-PT) the electromechanical coupling factor $k_{33}$ ~ 0.92 and the longitudinal piezoelectric constant $d_{33}$ of 1500 pC/N were reported [1]. Similarly, in Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO$_3$ (PMN-PT) $k_{33}$ ~ 0.90 and $d_{33}$ ~ 2000 pC/N has been reported [2]. With more evidence of single crystal relaxor ferroelectrics showing similar properties being reported, these materials seem to be ideal candidates for high bandwidth and high power density applications. However, the limitations for these materials are the low Curie temperature $T_C$, low ferroelectric rhombohedral $F_R$—ferroelectric tetragonal $F_T$ transition temperature $T_{RT}$, the low coercive field, and moreover, low mechanical quality factor $Q_m$ which limits the stability at high electric drive conditions. Heat generation sometimes limits the single crystal usage for practical transducer applications. This is the motivation of this research.
In single crystals, the properties change dramatically with the crystal orientation as reported in Reference [3]. Therefore, by using appropriate cut orientation of the single crystals the high power properties may be improved. Moreover, the above limitations can be mitigated by doping the single crystals with appropriate elements. In Reference [4], the mechanical quality factor improvement was observed in PZN-PT single crystals with Mn-doping. Therefore, in this paper we examine the loss properties in 0.70Pb(Mg_{1/3}Nb_{2/3})O_3-0.30PbTiO_3 single crystals as a function of (1) vibration mode (2) crystal orientation, and (3) doping.

The single crystal samples in k_{33} and k_{31} vibration modes were fabricated using Solid-State Crystal Growth (SSCG) technique [5]. Since the elastic, piezoelectric and dielectric properties of 0.71Pb(Mg_{1/3}Nb_{2/3})O_3-0.29PbTiO_3 were reported in Reference [6], we focus in this paper the loss/mechanical quality factor. The dimensions (12x4x4 mm³) of the 33- and (17x3x1 mm³) of the 31- vibration modes were chosen as per the IEEE standard guidelines [7]. Figure 4.1 shows the spontaneous polarization directions in this ferroelectric rhombohedral crystal (the coordinate axis is based on a cubic perovskite axis). The samples obtained were cut and electrically poled along the [001] and [011] directions, as shown in Figure 4.2. For each of the crystal orientations and vibrations as shown in Figure 2, un-doped as well as 1mol% Mn-doped and In-doped samples were fabricated.
Figure 4.1: Equivalent spontaneous polarization vectors of a ferroelectric rhombohedral PMN-PT crystal. The coordinate axes are based on cubic perovskite phase (not a rhombohedral ferroelectric phase).
4.2 Experimental Procedure

Generally used equations for the determination of the mechanical quality factor of a $k_{31}$ mode plate sample, as described in the IEEE standards [6], is obtained by the determination of the minimum impedance ($Z_m$) at the fundamental resonance frequency. $Q_m$ is given by the relation,

$$\frac{1}{Q_m} = 2\pi f \cdot Z_m \left( C_0 + C_1 \right) \left( \frac{f_B^2 - f_A^2}{f_B^2} \right)$$  \hspace{1cm} (4.1)
where,

\( f_A \) = fundamental resonance frequency,

\( f_B \) = anti-resonance frequency,

\( C_0 \) = shunt capacitance of the sample,

\( C_1 \) = series capacitance of the sample.

Note that Equation (4.1) is true only when \( Q_m \)'s for the resonance and antiresonance modes are the same, which does not happen in practical piezoelectrics. We reported the mechanical quality factor for resonance (\( Q_A \)) is higher than the mechanical quality factor for antiresonance (\( Q_B \)) in most of the cases in the previous paper [8], and the relationship between \( Q_A \) and \( Q_B \) can be provided by

\[
\frac{1}{Q_B} = \frac{1}{Q_A} + \frac{2}{1 + \left(\frac{1}{k_{31}} - k_{31}\right) \Omega_p^2} (\tan \delta' + \tan \varphi' - 2 \tan \theta')
\]

(4.2)

where \( \tan \delta', \tan \varphi' \) and \( \tan \theta' \) represent “intensive” dielectric, elastic and piezoelectric loss factors, respectively.

Another way of obtaining the quality factor is based on the frequency difference between two frequencies corresponding to the 3 dB below the resonance peak of resonance as well as anti-resonance peak. In piezoelectric materials, there exists resonance and anti-resonance exist as shown in Figure 4.3, which can be roughly understood as electrical short circuit condition and the latter at electrical open circuit condition respectively. The impedance curves were measured using HP4294 Impedance analyzer with an oscillation level of 0.5V.
Taking a narrow frequencies range close to the resonance and anti-resonance we can define the resonance quality factor ($Q_A$) and the anti-resonance quality factor ($Q_B$) as shown in Equation (4.3).

$$Q_A = \frac{f_A}{f_{A1} - f_{A2}}, \quad Q_B = \frac{f_B}{f_{B1} - f_{B2}}$$

(4.3)

where, $(f_{A1} - f_{A2})$ or $(f_{B1} - f_{B2})$, is the 3 dB bandwidth of the impedance curve.

The electromechanical coupling factors $k_{31}$ and $k_{33}$ were obtained using the following equations defined in the IEEE standards:

$$k_{31}^2 = \frac{\pi}{2} \frac{f_A}{f_B} \frac{1}{\pi f_A} - \tan \left( \frac{\pi f_A}{2 f_B} \right)$$

(4.4)
4.3 Results and Discussion

In this section, we summarize the properties of the single crystal samples. Figure 4.4 shows the measured quality factor and electromechanical coupling factor for the 31-vibration mode. Here, $Q_m$ is calculated using Equation (4.3), $Q_A$ is calculated using Equation (4.1) and $k_{31}$ is calculated using Equation (4.4) above. Note that in the horizontal axis of the figure, the notation PMN-PT [001] L[100] means that the length of the $k_{31}$ plate is cut along [100] axis. In Figure 4.5, the quality factor at anti-resonance is shown.
Figure 4.4: Mechanical quality factor and electromechanical coupling factor for $31$- vibration mode.
Figure 4.5: Anti-resonance mechanical quality factor for the 31- vibration mode.

Note that $Q_B$ seems to be 1.3 – 2 times higher than $Q_A$ in these single crystals, which concludes that the piezoelectric loss seems to be quite high compared with the dielectric and elastic losses; i.e., $2 \tan \theta' > (\tan \delta' + \tan \phi')$ in Equation (4.2).

Similarly, Figure 4.6 shows the mechanical quality factor and the electromechanical coupling factor for the k33 vibration mode. For the k33 vibration mode, the anti-resonance quality factor was not obtained experimentally, because the impedance of the k33 rod was higher than the dynamic range of the HP4194 impedance analyzer. Therefore, accurate and reproducible results could not be obtained in the k33 vibration mode at anti-resonance.
4.3.1 Effect of Vibration mode

Table 4.1 compares the quality factor at resonance $Q_A$ for the 33- and 31-vibration modes of the PMN-PT single crystals and PZT ceramics. It is clear that a large difference in quality factors exists for the two vibration modes. The 33- vibration mode $Q_A$ is almost twice that of the 31-mode. Table 1 also includes the $Q_A$ values for the 33- and 31- vibration modes of the PZT polycrystalline samples as reference, which do not show a significant difference between these two modes.
Table 4.1: Effect of vibration mode on quality factor

<table>
<thead>
<tr>
<th>Quality Factor, QA</th>
<th>33-mode</th>
<th>31-mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMN-PT [001]</td>
<td>330</td>
<td>146</td>
</tr>
<tr>
<td>In-PMN-PT [001]</td>
<td>270</td>
<td>113</td>
</tr>
<tr>
<td>Mn-PMN-PT [001]</td>
<td>516</td>
<td>240</td>
</tr>
<tr>
<td>‘Hard’ PZT (APC841)</td>
<td>1126</td>
<td>1183</td>
</tr>
<tr>
<td>‘Soft’ PZT (APC850)</td>
<td>100</td>
<td>75</td>
</tr>
</tbody>
</table>

4.3.2 Effect of Crystal Orientation

Figure 4.7 compares the resonance quality factor for the 31- and 33- vibration modes as a function of crystal orientation and doping. The crystal orientation effect on the quality factor of k33 and k31 mode is not pronounced.
4.3.3 Effect of Doping

From Figures 4.4, 4.5, 4.6 and 4.7, it can be seen that the mechanical quality factor for Mn-doped single crystals increases almost 3 times as compared to un-doped and In-doped. Note that the electromechanical coupling factor is still high, $k_{31}=0.83$ for [001] orientated crystal. This result is encouraging for the use of Mn-doped crystals for high power density applications.

4.4 Maximum Vibration Velocity of PMN-PT single crystals

Based on above loss analysis, it would be also useful to know the maximum vibration velocity for evaluating high power level, which could be achieved by these single crystals at resonance condition. For this experiment, the vibration velocity 31 vibration mode samples were measured using a laser fiber interferometer [Polytec, OFV 511], by applying sine wave signal to the sample at its resonance frequency at the same time observing the temperature rise at the nodal point. The vibration velocity (rms value) then was taken as the velocity at which the nodal point became 20°C above the room temperature. Figure 4.8 shows the comparison of the vibration velocity between piezoelectric single crystals and PZT ceramic samples measured as described above and having the same sample geometry. Typical “hard” and “soft” PZT samples ($k_{31}$ plate)
exhibit the maximum vibration velocities (rms value), 0.6 m/s and 0.15 m/s, respectively, while the PMN-PT crystals exhibit the intermediate values.

Figure 4.8: Vibration velocity of piezoelectric single crystals and ceramics.

The properties of PMN-PT single crystals doped with Mn with respect to the vibration velocity, as well as the loss properties described in the previous section, indicate that it is a semi-‘hard’ material with high piezoelectric constant and electromechanical coupling which is a good sign for high power applications.
References


8 Y. Zhuang, S.O. Ural, A. Rajapurkar, S. Tuncdemir, A. Amin and K. Uchino:

Chapter 5

Conclusions

Loss in PZT Piezoelectric Ceramics

In this thesis, the losses in piezoelectric ceramics are described in terms of the quality factor defined at resonance ($Q_A$) and anti-resonance ($Q_B$). Recommendation has been made for the sample holder based on the natural vibration direction of the piezoelectric sample. Theoretical derivation for the anti-resonance quality factor is discussed. Results of $Q$ values for ‘hard’ and ‘soft’ piezoelectric ceramic in $k_{33}$ and $k_{31}$ vibration modes were presented at low power as well as high power. Thus it is recommended to use anti-resonance frequency drive for driving in high power density applications like ultrasonic motors.

Loss in PMN-PT Single Crystals

In this paper, the loss properties of PMN-PT single crystals as a function of vibration mode, crystal orientation, and doping was discussed. It is observed that, the 33- vibration mode has a higher quality factor than the 31- mode. Besides, crystals orientation has no pronounced effect on the quality factor. Mn-doping significantly increased the quality factor of PMN-PT single crystals. Mn-doped single crystals exhibited ‘semi-hard’ loss properties while maintaining high electromechanical coupling, making them a promising candidate for high power density applications. In this research, we studied the properties for one particular dimension in each of the $k_{33}$ and the $k_{31}$ modes. It should be noted that the mechanical quality factor and other properties will change in PMN-PT relaxor ferroelectrics depending on the frequency.
Appendix A

Piezoelectric Constitutive equations

The commonly used variables to describe the state of a piezoelectric material are the stress $X$, strain $x$, electric field $E$ and electric polarization $P$ (or dielectric displacement $D$). We can express each electromechanical pair of the quantities in terms of others which gives the piezoelectric constitutive equations:

\[ X = c^E x - e'E \]  \hspace{1cm} (A.1)

\[ D = e^E x + e^X E \]  \hspace{1cm} (A.2)

\[ X = c^D x - h'D \]  \hspace{1cm} (A.3)

\[ E = -hx + \beta^X D \]  \hspace{1cm} (A.4)

\[ x = s^E X + d'E \]  \hspace{1cm} (A.5)

\[ D = dX + e^X E \]  \hspace{1cm} (A.6)

\[ x = s^D X + g'D \]  \hspace{1cm} (A.7)

\[ E = -gX + \beta^X D \]  \hspace{1cm} (A.8)
Bibliography


Vita

Aditya Rajapurkar received a Bachelor of Engineering degree in Electrical Engineering from the Maharaja Sayajirao University of Baroda, India in October 2006. This thesis is part of the requirements for his Master’s degree in Electrical Engineering with an emphasis on piezoelectric materials, actuators and transducers. Following is a list of publications, conference proceedings and presentations in his research area.

Publications


Conference Proceedings


Presentations

Mechanisms in Piezoelectric PZT Ceramics”, US Navy Workshop on Acoustic 
• A. Rajapurkar, S.O. Ural, S-H. Park, N. Bhattacharya and K. Uchino, 
“Anisotropic Loss Mechanisms in Piezoelectric Ceramics”, Material Science and 
Mechanisms in Piezoelectric Ceramics”, 50\textsuperscript{th} ICAT/JTTAS Smart Actuator 
• A. Rajapurkar, S.O. Ural, S-H. Park, N. Bhattacharya and K. Uchino, 
“Anisotropic Loss Mechanisms in Piezoelectric Ceramics”, Material Science and 
• A. Rajapurkar, S.O. Ural, S-H. Park, N. Bhattacharya and K. Uchino, 
“Anisotropic Loss Mechanisms in Piezoelectric Ceramics”, US Navy Workshop 