Structural , Dielectric and Electrical Properties of Lead Zirconium Titanate (PZT) Ceramics

Abstract

Structural properties of the compounds were examined using an X-ray diffraction (XRD) technique to confirm the formation of phase at different temperature. The electrical current response for equivalent circuit of PZT, resonant frequencies, antiresonant frequencies and mechanical quality factor studied by numerical simulation and compared with experimental results. The dielectric properties of PZT ceramics in the temperature range 50 -200 C^0 were measured. The effect of grain size of the PZT on the dielectric constant and dielectric loss were investigated. As the grain size increased, the maximum dielectric constant increased. The dielectric study with frequency at different temperature in the frequency range 1 to 5 MHz shows that dielectric constant decreases with increasing frequency. Loss factor does not vary with frequency but it becomes independent of higher frequency range

Keyword, PZT; dielectric loss; quality factor; electric impedance;grain size

1. Introduction

Lead Zirconate titanate (PZT) is an important and promising ferroelectric material used in applications like sensors, ultrasonic transducers and electro-optic devices for data storage[1,2]. Lead zirconate titanate (PZT) are representative perovskite ferroelectric and piezoelectric prototypes because of their excellent electrical properties [3, 4]. However, the best dielectric and piezoelectric properties of PZT have been found near MPB [**5**]. K.K. Shung wrote a review article on Piezoelectric materials for high frequency medical imaging applications [6]. Piezoceramic material of particular interest, in this work, is the PZT-5A mixture, since it has been used extensively in ultrasonic probes. These materials offer a high thickness coupling coefficient, which corresponds to higher sensitivity. The relatively low clamped dielectric constant of PZT-5A makes it suitable for large aperture pulsed devices [7]. Uchino et.al [8] describes the basic formulae for the loss in piezoelectric materials in terms of electrical and mechanical dissipation factors. Losses in piezoelectric materials can be divided into three different mechanisms: dielectric, mechanical and piezoelectric losses. 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 is inversely proportional to the mechanical loss factor [9]. Mezheritsky [10] derived analytical expressions for the quality factors observed at resonance and anti resonance. The effects of grain size and dc. electric field on the dielectric properties of the Barium strontium titanate ceramics has been investigated by Tang et al [11]. Banerjee et al [12] also examined The Influence of Particle Size of PZT-epoxy-Al composites on the Dielectric Properties.

In this paper, The disk type ceramic specimen based on the PZT-5A material were fabricated using conventional methods. It had 20-mm diameter and 2-mm thickness. The resonant frequency, antiresonant frequency and mechanical quality factor studied by numerical simulation and compared with experimental results. The piezoelectric constants for PZT5A are calculated. The dielectric properties of PZT5A ceramics in the temperature range 50 -200 C^0 were measured by the effect of grain size.

2. Dynamic Behavior of Piezoceramic Transducer

When the piezoceramic disk exposed to an alternating electric field it periodically oscillates in accordance with the applied field. In this case its behavior can be described by the variation of the electrical impedance with frequency of oscillation. There are two approaches to obtain the impedance of such resonator as electrical circuit impedance each of which was derived from the piezoceramic constitutive relations. An easier approach based on the measurement of resonance f_r and antiresonance f_a frequencies of unloaded, unclamped disk, which resonates in the thickness mode. In such case piezoceramic resonator disc can be considered as a pure electric reactance its impedance given as;

$$Z_{Elec.} = \frac{t}{j\omega A \varepsilon_{33}^{s}} \left[1 - \frac{K_t^2 \tan(\omega / 4 f_r}{\omega / 4 f_r} \right] \qquad (1)$$

Where t,A,ω , K_t^2 are thickness, area of the disk, angular frequency and coupling coefficient. ε_{33}^{s} is dielectric permittivity at constant strain (clamped). Therefore another theoretical prediction of such impedance may be abstracted from 1D thickness mode resonator model [13], since the Laplacian transform of the resonator electrical impedance Z(s) given in the form;

$$Z_{elec.} = \frac{1}{sC_0}\left[1 - \frac{k_t^2}{sT}\left\{\frac{K_f T_f}{2} + \frac{K_B T_B}{2}\right\}\right] \tag{2}$$

Where the functions K_f and K_B are defined as,

$$K_f(s) = \frac{\left(1 - e^{-sT}\right)\left(1 - R_B e^{-sT}\right)}{\left(1 - R_f R_B e^{-2sT}\right)} \tag{3}$$

$$K_B(s) = \frac{\left(1 - e^{-sT}\right)\left(1 - R_f e^{-sT}\right)}{\left(1 - R_f R_B e^{-2sT}\right)} \tag{4}$$

where, s is the Laplacian complex variable, T is the one way propagation time, T_F and T_B are the transmission coefficients from the front and back faces of the disc. R_F and R_B are the reflection coefficients from the front and back faces of the disc and C_0 is the clamped capacitance of the transducer. Equation(2) will be used to predict the electric impedance as a function of frequency. 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. Generally, in the data sheet available for commercial piezoelectric materials the mechanical quality factor (Q_m) and the dielectric dissipation factor (tan ∂) are provided for the loss characteristics. In this case Qm is calculated near the resonance frequency from a piezoelectric disc in planar vibration mode [14-15]. 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_r) and anti-resonance frequency (f_a), the capacitance and the dissipation factor in the desired frequency range are required to determine the material constants. The mechanical quality factor Q_m is obtained from the determination of the minimum impedance Z_r at the fundamental resonance. This measurement is accomplished by substitution of a variable resistance for the test specimen at f_r. Q_m is given by the follow relation.

$$\frac{1}{Q_m} = 2\pi f_r Z_r C_f \frac{f_a^2 - f_r^2}{f_a^2} \quad (5)$$

C_f is the capacitance measured under the low frequency condition (1 kHz). The electromechanical coupling factors k_{31} and k_{33} were obtained using the following equations defined in the IEEE standards:.

$$k_{33}^2 = \frac{2f_r}{\pi f_a} \tan(\frac{\pi(f_a - f_r)}{2f_a} \quad (6)$$

$$\frac{1}{s_{33}^D} = 4\rho f_a^2 l^2 \quad (7)$$

$$s_{33}^E = s_{33}^D / (1 - k_{33}^2) \quad (8)$$

$$d_{33} = k_{33}\sqrt{\varepsilon_{33}^T s_{33}^E} \quad (9)$$

The planar coupling factor k_p is obtained from the fundamental frequency f_r and f_a of a disk as follows

$$k_p^2 = \frac{1}{p}\frac{f_a^2 - f_r^2}{f_a^2} \quad (10)$$

$$p = \frac{2(1+\sigma^E)}{\eta_1^2 - 1 - (\sigma^E)^2} \quad (11)$$

Where σ^E is Poisson's ratio, and μ_1 is the lowest positive root of $(1+\sigma^E)J_1(\eta) = \eta J_0(\eta) = \eta J_0(\eta)$. Where J_1 is the Bessel function of the first kind.

3. Experimental approach

Figure (1) shows the Piezoceramic disk of diameter 10 mm (axes 1 and 2) which are much greater compared to its thickness t = 2.0 mm (axis 3). This choice establishes the following conditions $E1 = E2$ = zero, and $\partial E3/\partial y = \partial E3/\partial z$ = zero. The constitutive relations of piezoceramic plate, which resonates in the one mode, have also led to that $D1 = D2$ = zero. The pointing vector **E** × **H** (*Maxwell equations*) of such resonator also zeroes i.e. there is no electromagnetic radiation is produced by this piezoceramic resonator, only mechanical radiation [16]. A resonator satisfying these restrictions can, therefore, operate in a thickness mode configuration[17]. The disk type ceramic specimen based on the PZT-5a material were fabricated. It had 20-

mm diameter and 2-mm thickness. The PZT desk were sintered at 50–200 °C for 3 h. Various grain sizes of PZT ceramics were obtained. X-ray diffraction patterns of PZT disc in the range of 2θ = 20-60° are shown in Fig. (2a) at 50 C^0 and (b) at 200 c^0. It can be found that the PZT crystal phase was grown from amorphous to perovskite phase was observed in the XRD patterns. The X-ray diffraction patterns matched with JCPDS card: 33-0784. The standard sample of NaCl is used to correct the measured diffraction for instrumental contribution. According to the width decomposition method the calculated volume grain size of the sample was equal 15.10 μm at 50 c^0 while it increased to 21.23 μm at 200 C^0**.**

3.1. Dielectric behaviour

Experimental impedance data from a disk of PZT has been used to measure the dielectric constant and dielectric loss properties of this material. Silver paste was coated to form electrodes on both sides of the sintered ceramic specimen for dielectric measurements. The dielectric constant and loss tan ∂ of the sample was measured using a LCR meter from 0.1 to 5 MHz. The impedance curves were measured with an oscillation level of 0.5 V. Fig.(3) shows the measured and predicted results of electric impedance for piezoceramic disk. It is clear from fig.(3) that the predicted and measured results for resonance (f_r) and anti-resonance frequency (f_a) equal to 0.90 ,0.955 MHz and 1.10, 1.10 MHz respectively. The frequency dependence of dielectric constant and loss tan (∂) of PZT sample with different temperature annealing of 50 C^0, 100 c^0 and 200 c^0 as shown in figs. 4(a) and 4(b). It is clear from these figures that the dielectric constant increases from 7134 to 10311 with an increase in grain size from 15.1 to 21.2 μm at 3MHz. Also, the dielectric loss increases from 0.033 to 0.048 at 3MHZ. Table(1) shows the computational results of Q_m, k_{33}, d_{33} and k_p by the equations described above. These results show that predicted results are good agreement with experimental results.

4. Conclusion

In this paper, the losses in piezoelectric ceramics are described in terms of the quality factor defined at resonance and anti-resonance. The measured resonant frequencies agree with the computational result and calculated the PZT constants. The dielectric study with frequency at different temperature in the frequency range 1 to 5 MHz shows that dielectric constant decreases with increasing frequency. Loss factor does not vary with frequency but it becomes independent of higher frequency range . Grain size effects of PZT ceramic on the dielectric properties were investigated. As the grain size increased, the maximum dielectric constant increased. The results can be used to careful piezoelectric transducer design to arrive at an adequate compromise between quality factor and sensitivity.

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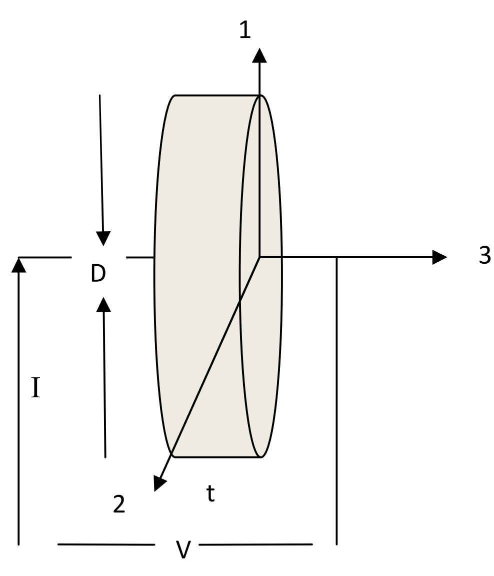

Fig.(1) PZT disc under compression force acting on direction 3

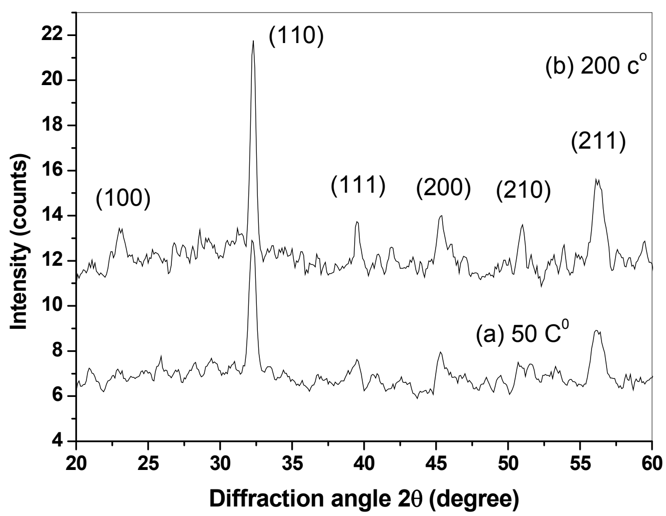

Fig.(2) XRD diffraction pattern of the PZT at: (a) 50 C^{0} and (b) 200 C^{0}

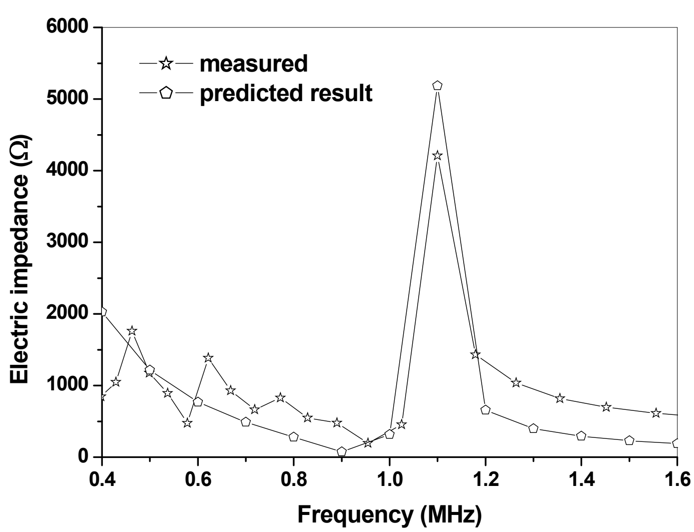

Fig.(3) electrical magnitude spectrum for the disk of PZT

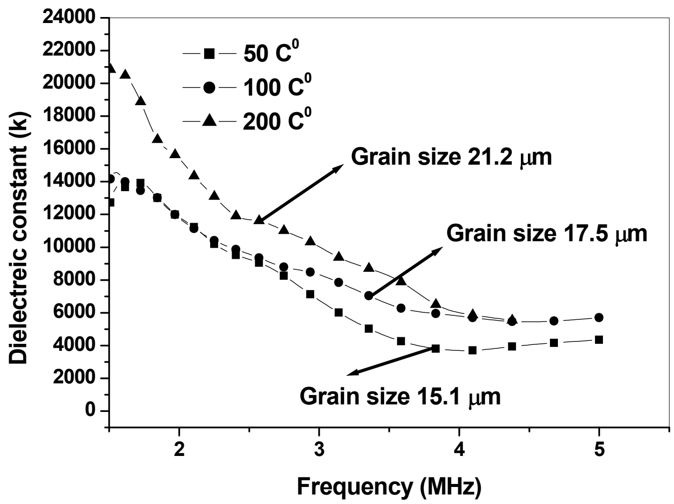

Fig.4(a) Change of dielectric constant with frequency of PZT

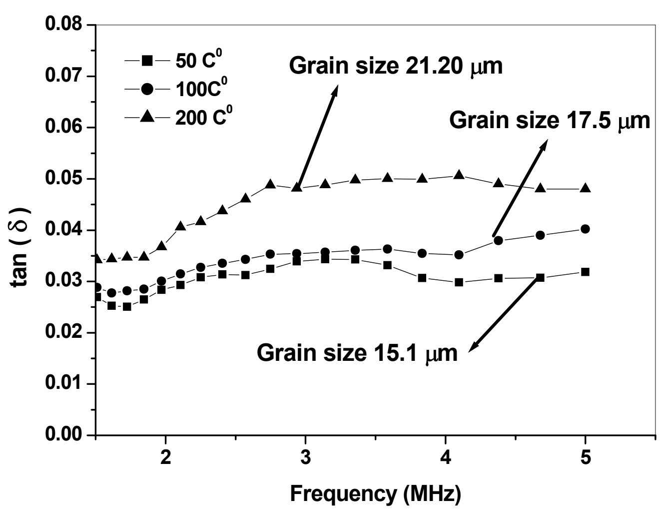

Fig.4(b) Change of dielectric loss with a frequency of PZT

Table (1) computational result of PZT parameters.

PZT parameters	Predicted results	Measured results
f_r (MHz)	.900	0.955
f_a (MHz)	1.100	1.100
Q_m	75.28	70.58
k_{33}	0.663	0.632
d_{33} (10^{-12} C/N)	276.6	258
k_p	0.413	0.307