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Effect of Thermal Shocking and Quenching on the Degradation Behaviour of a Thin PZT Disc

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Abstract. Thin lead zirconate titanate discs were subjected to thirty five thermal shocks from two different temperatures in deionized water and their relative dielectric constant, coupling factor and impedance values were measured with a view to investigating the behaviour of thin piezoelectric (PZT) discs at frequency of maximum and minimum impedance. Noticeable differences were observed in the electrical properties of the material, probably due to the change in dipole lengths and their orientations during thermal shocking. The results can be useful in modeling and designing of smart components for predicting their behaviour during such expected shocking conditions prior to fabrication.

Keywords: piezoelectric material, thermal shock, deionized water, dielectric constant, impedance, PZT

Introduction

Piezoelectric materials are used in various electromechanical applications where they are influenced by various cyclic loadings. Thermal cycling or thermal fatigue in most electronics materials may cause degradation in their internal characteristics. Thermal fatigue test methods include quench method and repeated heating method for thermal shocks which have been earlier discussed (Lamon and Pherson, 1991; Lamon, 1981). Influence of temperature on the electromechanical and fatigue behaviour of piezoelectric ceramics has been studied by Wang *et al*. (1998). Temperature gradient is developed due to sudden change in temperature in the ceramic materials and therefore, thermal stress is generated. Effect of thermal shocks has been studied by developing newly designed equipment. There are various popular thermal shock methods available in ascending and descending orders. Some of them popular for ascending thermal shocks, include hot jet gas method, high power radiation, melt immersion test, ribbon test method and high power laser heating method. Similarly, various test methods for descending thermal shocks are quenching in water, fluidized bed or a cold air jet impinging on hot discs; quenching in contact with huge brass rods and indentation method have been mentioned by Panda *et al*. (2002). Earlier, thermal shocks in a plate of finite thickness had been attempted. Thermal shock and thermal fatigue of ferroelectric thin films were investigated by Zheng *et al*. (2005). In all of the above methods, the water quenching method is mostly used for thermal shock tests in which samples are heated to a particular temperature and then quenched in water bath. Fatigue studies show that material degradation of PZT ceramics are strongly influenced by temperature. Lead zirconate titanate ceramics show decrease in the dielectric constant and the resonance frequency when subjected to thermal shocks. Importance of temperature stability for dielectric constants and resonance frequencies have been discussed by Lee and Kim (2005). Earlier thermal shock resistance of the materials was evaluated by water quenching. Degradation of various properties of the piezoelectric devices in the presence of water and AC voltage was investigated by Xiang *et al*. (2007). They concluded that water is an important cause of degradation of piezoelectric (PZT) ceramics. However, limited work has been published on the effect of thermal shocking, quenching and on the degradation behaviour of thin piezoelectric ceramic discs. In this study, the degradation phenomenon of thin PZT ceramic disc have been investigated when exposed to repeated heating and quenching cycles below its curie temperature.

Materials and Methods

Lead zirconate titanate piezoelectric discs, nickel electroded on major faces, 0.191 mm thick and 12.7 mm in diameter, were used for the experimentation. The thin piezoelectric ceramic discs were heated at the heating rate of 9 °C/sec up to 100 °C, and 150 °C, using a thermal chamber and then quenched in deionized water at a temperature of about 20 °C. For all thermal cycling and quenching experiments, 2 PZT test samples were used and subjected to identical conditions. The temperature of the PZT samples was recorded using a spring loaded thermocouple and data acquisition system attached directly to the samples. In order to observe degradation phenomenon of the PZT ceramic, the capacitance, dissipation factor and impedance were measured at a frequency of 1 kHz at the start and after every five heating and quenching

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cycles. Data was collected for a total of thirty-five thermal shocks and their relative frequencies of maximum and minimum impedance were observed between 100 kHz and 200 kHz. The capacitance and impedance at these frequencies were recorded using impedance analyzer and dielectric test fixture (model 1645 B). The fixture was attached to an LCR meter and impedance analyzer 4294 A which uses a 4 pair terminal measurement configuration. The values of capacitance measured with impedance analyzer were used to calculate dielectric constant (K_3^T and effective coupling factor (K_{eff}) by using the following equations from IEEE Standard 177 (IEEE Standard, 1976) and Moulson and Herbert (2005):

$$K_3^T = \frac{t_a \times C_p}{A \times \epsilon_0}$$

Effective and transverse coupling factors (K_{eff}) were determined by using the following relationships:

$$K_{eff} = \mathrm{SQRT}\,(f_n^{\,2} - f_m^{\,2}) / f_n^{\,2}$$

$$K_{31} = \mathrm{SQRT}\,(\Psi/(1+\Psi)),$$

where $\Psi = \pi/2\,(f_n/f_m) \times \tan|\pi/2 \times (f_n - f_m) / f_m|$

Abbreviations used are as follows:

f_m =	frequency of maximum impedance	[Hz]
f_n =	frequency of minimum impedance	[Hz]
C_p =	equivalent parallel capacitance	[F]
t_a =	average thickness of testing material	[m]
A =	area of guarded electrode	[m^2]
K_3^T =	dielectric constant	
K_{eff} =	effective coupling factor	
K_{31} =	coupling factor with transverse excitation	
ϵ_0 =	permittivity at free space (8.854×10^{-12})	
Ψ =	phase angle	

Results and Discussion

The changes in dielectric constant and coupling factor were measured as a function of frequency of maximum and minimum impedance (f_m and f_n, respectively). Increase in the value of the capacitance of the as-received PZT ceramic was observed to be 5.8×10^4 pF which gradually decreased with increasing thermal cycling (100 °C - 20 °C) to 1.72×10^4 pF. A corresponding change in the f_m was observed with a value of 160 kHz for the PZT sample at the start and then the value decreased to 116.5 kHz. This represented a 28% decrease in the f_m after the ceramic was thermal cycled. A similar change was observed for the f_n which decreased from 165.5 kHz for the as-received to 153.3 kHz after 35 thermal cycles. For the thermal cycling (150 °C - 20 °C) change in f_m was observed from 160.2 kHz to 141 kHz and from 165.175 kHz to 157.5 kHz in f_n. Change in dielectric constant and coupling factor for thirty five shocks in deionized water has been tabulated in Table 1.

Figure 1 indicates the value of capacitance directly measured by impedance analyzer for the unshocked discs at frequency of maximum impedance. Discs were shocked in deionized water from 100 °C to 20 °C for thirty five shocks when their capacitance value decreased from 58.041 nF to 17.237 nF (Fig. 1 and 2). Interestingly, PZT discs shocked from 150 °C to 20 °C showed a less decrease in capacitance value after having thirty five shocks. In this case capacitance value at frequency of maximum impedance decreased to 24.189 nF (Fig. 3).

A comparison of the graphical output for dielectric constant for the PZT samples before thermal cycling and then after exposing the ceramic to thirty five heating and quenching shocks is shown in Fig. 4. Dielectric constant remains independent when measured at 1kHz. The dielectric constant is an

Table 1. Change in dielectric constant and coupling factor for two different thermal shocking conditions

	Shocking from 100 °C to 20 °C				Shocking from 150 °C to 20 °C			
Shocks #	K_3^T at 1kHz	K_3^T at f_m	K_{eff}	K_{31}	K_3^T at 1kHz	K_3^T at f_m	K_{eff}	K_{31}
0	1853	9888	0.255	0.279	1869	10462	0.23	0.26
5	1891	9481	0.313	0.34	1915	8532	0.27	0.29
10	1911	7726	0.324	0.352	1923	7005	0.3	0.32
15	1930	6392	0.306	0.333	1932	4366	0.32	0.35
20	1918	5291	0.364	0.393	1954	3926	0.34	0.37
25	1922	4790	0.427	0.457	1976	3504	0.36	0.38
30	1928	3450	0.621	0.644	1976	3912	0.41	0.41
35	1928	2935	0.651	0.671	1976	4121	0.44	0.47

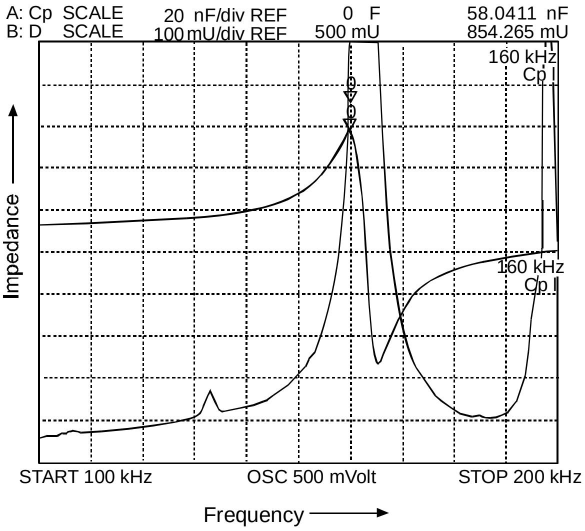

Fig. 1. Value of capacitance for un-shocked disc w.r.t. frequency (100 kHz-200 kHz); capacitance in nF at frequency of maximum impedance.

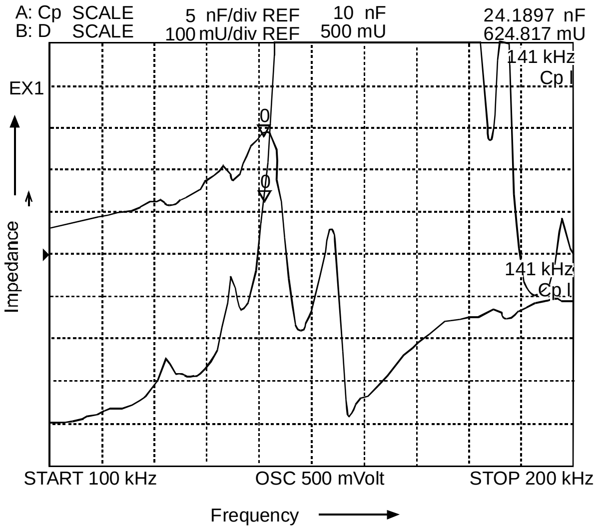

Fig. 3. Value of capacitance after thirty five shocked w.r.t. frequency (100 kHz-200 kHz); capacitance in nF at frequency of maximum impedance when shocked from 150 °C - 20 °C.

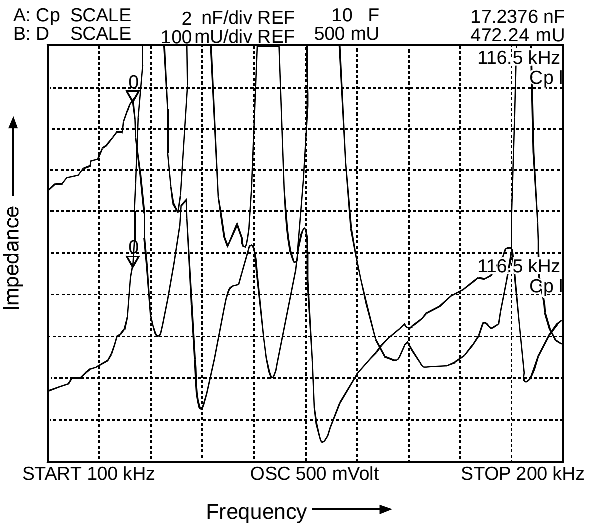

Fig. 2. Value of capacitance after thirty five shocks w.r.t. frequency (100 kHz-200 kHz); capacitance in nF at frequency of maximum impedance when shocked from 100 °C - 20 °C.

intrinsic property of the ceramic material and the results show that this value decreased with increasing thermal cycles at f_m and *vice versa*. The relative difference in the frequencies of the maximum and minimum impedance values depends on the material coupling factor and the resonator geometry (i.e., dimensions of the ceramic PZT sample). For this reason, quantities known as the effective coupling factor (K_{eff}) and the transverse excitation factor (K_{31}) were calculated and compared as a function of the number of thermal shocks (Fig. 5). It was found that both the values of K_{31} and K_{eff} increased with increasing thermal cycles to which the PZT ceramic was exposed.

The change in modulus of impedance for two different shocking conditions were evaluated (Fig. 6). It can be seen that the modulus of impedance for both f_m and f_n, when shocked from 100 °C to 20 °C, increased whereas for the other conditions, it started decreasing after twenty five shocks. Another interesting result is that when impedance at f_m and f_n increased, the difference became larger at later shocks. Decrease in dielectric constant in thermal shocking is the expected normal behaviour. Various coupling factors were close to each other but a noticeable change was observed due to shocking and quenching effect. All changes may have occurred due to change in dipole moments and their expected random orientations. This reorientation may change the length of dipoles, due to which specimen undergoes a change in its piezoelectric properties.

The change in f_m and f_n causes the change of mechanical quality factor. This response of the material can be utilized in designing of oscillators. It is observed that the difference in these two stated frequencies (f_m and f_n) is small as compared to their impedance peaks during thermal shocking. The

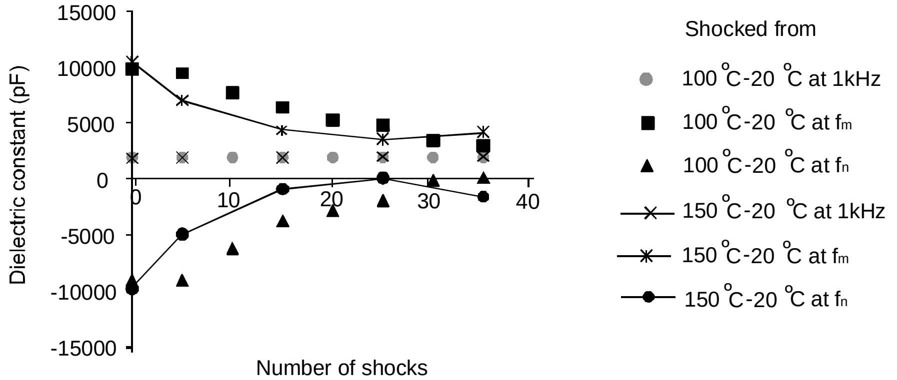

Fig. 4. Change in dielectric constant against number of shocks, at frequency 1kHz and frequencies of maximum and minimimum impedance.

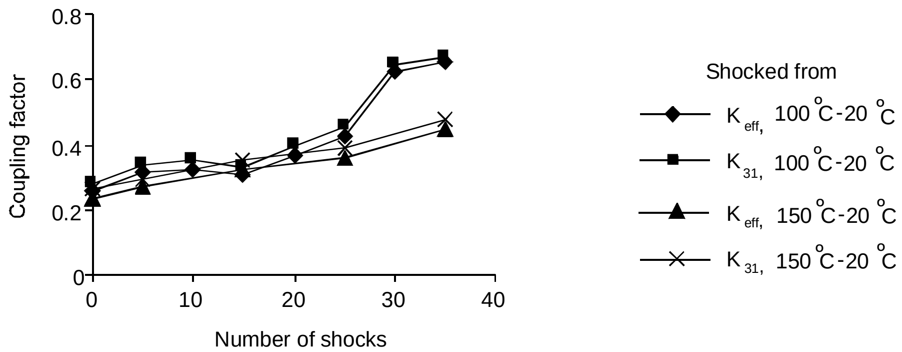

Fig. 5. Change in coupling factor (K_{31}, K_{eff}) against number of shocks from 100 °C - 20 °C and from 150 °C - 20 °C in deionized water.

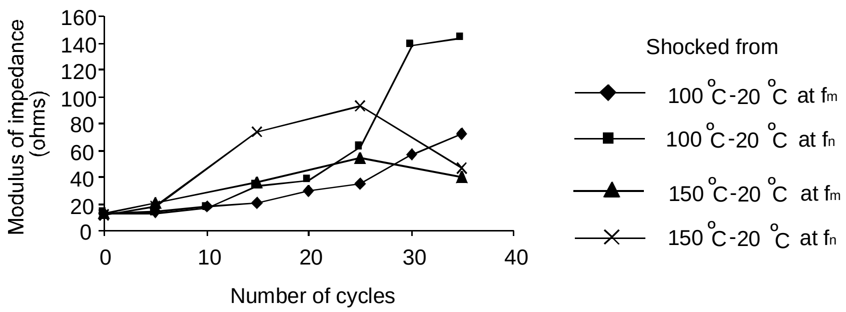

Fig. 6. Change in modulus of impedance (|Z|) against number of shocks from 100 °C - 20 °C and from 150 °C - 20 °C in deionized water.

results suggest that the PZT ceramics suffer a noticeable change in polarization when exposed to repeated heating and quenching cycles, well below the curie temperature (350 °C) for the PZT ceramic. It is thought that significant depolarization of the PZT ceramic occurs due to the disorientation of the ferroelectric domains and this reorientation is affecting the critical piezoelectric properties by thermal shocking and quenching. The behaviour is normal but the number of peaks increases due to expected change in length and reorientation of dipoles. Development of dipole moments