A Novel Method for Measurement of Equivalent Circuit Components of Piezoelectric Materials by Using Impedance Spectroscopy

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Abstract
A piezoelectric material is represented by an equivalent circuit model which usually has a series LCR circuit connected in parallel to another capacitance. Measurement of the values of the components by using impedance spectroscopy and complex non-linear least squares is presented. The method is demonstrated by taking an example and comparing the calculated values of the components with the assigned values.

1. Introduction

Piezoelectrics in single crystalline, ceramics or polymeric form are very important class of technological materials having wide variety of applications such as piezoelectric resonant elements used in frequency standards, electric field sensors, electromechanical transducers, chemical and biological sensors, energy harvesters, gas igniters, impact sensors etc[1- 3]. In majority of applications the material prepared in certain shapes and forms suitable to yield maximum transduction is attached to some other system. In order to achieve reproducibility and optimum functioning and power transfer detailed knowledge of electrical behavior of the piezoelectric material is desired. This is greatly facilitated by representing the piezoelectric by a suitable equivalent circuit model and determining the values of the components used in the model [4-9]. These values can be estimated by observing the resonance and anti-resonance frequencies and plotting the total admittance vs frequency graphs as well as vector admittance diagrams [10, 11]. Methods have been proposed for obtaining the values of the material properties of the resonators by using curve fitting approach and representing the lumped circuit elements by complex quantities to include losses [6]. However straight forward ways to decide upon the initial guesses for these values are not available. In this paper a method based on impedance spectroscopy is presented for obtaining the values of the components of the equivalent circuit model. The approximate values of the components are obtained from the spectroscopic and complex plane plots and are further refined by using Complex Nonlinear Least Squares (CNLS) procedure. All the components are taken as real. The basics of impedance spectroscopy are explained in the next section followed by expressions for immittance (impedance, admittance, electric modulus and permittivity) of the most accepted model of piezoelectrics for ready reference and method for obtaining the values of the components.

2. Brief Description of Impedance Spectroscopy

The technique of Impedance Spectroscopy can be easily understood by considering the impedance behaviour of a polycrystalline electronic ceramics. A polycrystalline ceramic comprises a large number of small crystallites called grains joined in random orientation. The inter- grain region, called grain-boundary (gb) has strained bonds due to mismatch in the grain orientations. The properties of the grain boundaries are therefore different from those of the grains. This fact gives rise to some very interesting and useful properties which are exploited in the commercial devices [1]. New horizons of applications are being witnessed and contemplated by reducing the grain size to nanometer range giving rise to the so-called nanomaterial. For the fabrication of a device conducting leads are connected to the ceramic product having a conducting coating or conducting layer. The flow of charge from one lead to the other would therefore be influenced by the behaviour of grain, grain boundary, and electrode. An electronic component thus can be viewed as a system, comprising grains, grain-boundary and contact electrode. In order to ensure reproducibility and to develop materials having tailor made properties, these so called grain, grain boundary and electrode contributions must be separated out and monitored suitably. The method of Complex Impedance Analysis or Impedance Spectroscopy [12] has emerged as a very powerful tool for this. The availability of high quality impedance analyzers working over extended frequency regions has made impedance spectroscopy an extremely popular field in the last few years and increasing trend is being witnessed for its wide spread applications in various areas[13, 14] including biosciences, medical, agriculture , engineering , electrochemistry etc. It is evident from the enormous amount of literature available on the internet.

In this method experimentally measured values of complex impedance ($Z^* = Z' - j\, Z''$, $j = \sqrt{-1}$) are plotted as a function of frequency or on complex plane i.e., Z'' vs Z'. These plots show special features depending upon relative contributions from grain, grain boundary and electrode polarization processes. For the case when these processes have widely separated time constants distinct semicircular arcs are obtained and when they have time constants close to each other depressed looking semicircular arcs or distorted arcs are obtained. A suitable model is chosen to represent the material- electrode system and the values of the components used are obtained from the intercepts of the arcs on the Z' axis, peaks in the plots and the values of the frequency where Z'' shows a maximum. Once the model has been established the effect of processing parameters, additives, electrode etc. can be monitored by observing the changes occurring in the values of the components. The choice of a suitable equivalent circuit is a difficult process as many equivalent circuit models can give rise to the same simulated behaviour. Therefore simple model circuits may be preferred to begin with. A parallel RC circuit has one time constant and thus can be used to represent one polarization/ charge transfer process. A general practice is to represent one polarization process by one parallel RC combination to start with and then put several such RC's in series each one representing a charge transfer process. The number of these RC's may be intuitively chosen to be equal to the number of polarization / charge transfer processes that seem to be present in the system. Thus a simple model for an electronic ceramic could be a combination of three parallel RC circuits connected in series representing grain, grain- boundary and contact electrode processes. The choice of the most appropriate circuit may be made by taking into consideration the qualitative behaviour and the microstructure of the material. This choice is prompted by comparing the experimental plots with the simulated patterns for various possible models [12,15,16].

The electrical behavior of a system can be expressed in terms of interrelated functions known as impedance ($Z^* = Z' - j\, Z''$), admittance ($Y^* = (Z^*)^{-1} = Y' + j\, Y''$), permittivity ($\varepsilon^* = (j\, \omega\, C_0\, Z^*)^{-1} = \varepsilon' - j\, \varepsilon''$) and modulus ($M^* = (\varepsilon^*)^{-1} = j\, \omega\, C_0\, Z^* = M' + j\, M''$) where $j = \sqrt{-1}$ and $\omega = 2\pi f$, f being the frequency of the ac excitations and C_0, the capacitance of the empty cell used to house the sample. Due to specific relationship between these broadly termed immittance functions they are used to extract information about the component used in the models. It has been reported that study of the ceramic system based on the information conveyed by only one of these four functions does not suffice and two functions viz impedance (Z^*) and modulus (M^*) or admittance (Y^*) and permittivity (ε^*) should be looked at [12, 17]. Computer programs are used for this purpose using graphics.

For any dielectric measurement the specimen is placed between the two electrodes. The total capacitance and the losses are governed by the dielectric constant and the loss factor of the material. The simplest equivalent circuit model for this electrode-specimen system would be a capacitance C with a resistance R in series or parallel as shown in Fig.1. Such a circuit will have a single time constant RC and thus can be thought of as representing a polarization process with relaxation time RC.
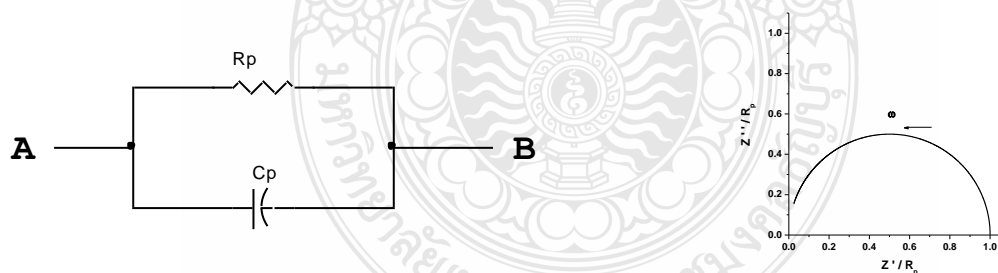

Fig.1 A resistor and capacitor connected in parallel and normalized Z'' vs Z'
plot for a parallel RC circuit

The value of impedance in parallel combination is given by the equation

$$Z_{AB} = \frac{R_p}{1 + \omega^2 C_p^{\,2} R_p^{\,2}} - \frac{j\omega C_p R_p^{\,2}}{1 + \omega^2 C_p^{\,2} R_p^{\,2}} \qquad \ldots(1)$$

Writing $Z_{AB} = Z' - j\, Z''$ we have

$$Z' = \frac{R_p}{1 + \omega^2 C_p^{\,2} R_p^{\,2}} \qquad\qquad Z'' = \frac{\omega C_p R_p^{\,2}}{1 + \omega^2 C_p^{\,2} R_p^{\,2}}$$

It can be shown that Z' and Z'' satisfy the relation $(Z' - R_p/2)^2 + Z''^2 = (R_p/2)^2$

This is equation of a circle with center at $(R_P/2, 0)$ and radius $(R_P/2)$ in a Z" vs Z' plot. As mentioned earlier, impedance is a function of frequency, so if we measure Z" and Z' at different frequencies and plot the graph between Z" and Z', we should get a circle. As Z" is a positive quantity we would get a semicircle as shown in Fig.1. This plot is known as complex impedance plot. From Eq. (1) we see that at $\omega = 0$, $Z' = R_P$ and $Z" = 0$. This implies that the point $(R_P, 0)$ in the complex impedance plot which is the intersection of the semicircle with the Z'- axis corresponds to the zero frequency. Therefore if ac measurements are done, i.e., Z' and Z" are experimentally measured at different frequencies then the dc value of resistivity can also be obtained from the complex impedance plot. Similarly we find that M' and M" satisfy the equation $(M' - C_0/2C_p)^2 + M"^2 = (C_0/2C_p)^2$ and the high frequency intercept would yield the value of $C_0/2C_p$.

3. The Equivalent circuit model of a piezoelectric
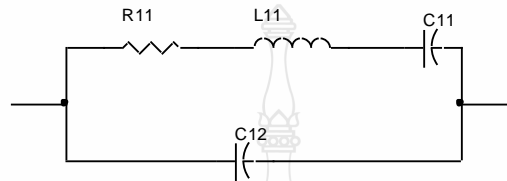The most accepted model of a piezoelectric resonator [3,18] is shown in Fig. 2

Fig. 2 Electric Equivalent of piezoelectric resonator

where R_{11}, L_{11}, C_{11} and C_{12} are resistance, inductance and capacitance respectively. The real and imaginary part of immittance function are

$$Z' = \frac{R_{11}}{\left[1 + \omega C_{12}\left(\frac{1}{\omega C_{11}} - \omega L_{11}\right)\right]^2 + \left[\omega C_{12} R_{11}\right]^2} \qquad \ldots(2)$$

$$Z" = \frac{\left(\frac{1}{\omega C_{11}} - \omega L_{11}\right) + \omega C_{12}\left[R_{11}^2 + \left(\frac{1}{\omega C_{11}} - \omega L_{11}\right)^2\right]}{\left[1 + \omega C_{12}\left(\frac{1}{\omega C_{11}} - \omega L_{11}\right)\right]^2 + \left[\omega C_{12} R_{11}\right]^2} \qquad \ldots(3)$$

$$Y' = \frac{R_{11}}{R_{11}^2 + \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)^2}, \qquad Y" = \frac{-\left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)}{R_{11}^2 + \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)^2} + \omega C_{12} \qquad \ldots(4)$$

$$M' = \omega C_0 \left(\frac{\omega C_{12} R_{11}^2 - \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right) + \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)^2 \omega C_{12}}{\left[1 - \omega C_{12}\left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)\right]^2 + \left[\omega C_{12} R_{11}\right]^2}\right) \qquad \ldots(5)$$

$$M" = \frac{\omega C_0 R_{11}}{\left[1 - \omega C_{12}\left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)\right]^2 + \left[\omega C_{12} R_{11}\right]^2} \qquad \ldots(6)$$

$$\varepsilon' = \frac{C_{12}}{C_0} - \frac{\left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)}{\omega C_0\left[R_{11}^2 + \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)^2\right]} \qquad \varepsilon" = \frac{R_{11}}{\omega C_0\left[R_{11}^2 + \left(\omega L_{11} - \frac{1}{\omega C_{11}}\right)^2\right]} \qquad \ldots(7)$$

The limiting values of the immittance functions at very low and very high frequencies are

$$Z'\big|_{\omega\to0} = \left(\frac{C_{11}}{C_{11}+C_{12}}\right)^2 R_{11}\ , \ Z'\big|_{\omega\to\infty} = 0\ , \qquad Z''\big|_{\omega\to0} = \infty\ , \ Z''\big|_{\omega\to\infty} = 0$$

$$M'\big|_{\omega\to0} = \left(\frac{C_0}{C_{11}+C_{12}}\right),\ M'\big|_{\omega\to\infty} = \frac{C_0}{C_{12}}\ , \qquad M''\big|_{\omega\to0} = 0\ ,\ M''\big|_{\omega\to\infty} = 0$$

$$Y'\big|_{\omega\to0} = 0\ ,\ Y'\big|_{\omega\to\infty} = \frac{1}{R_{11}}\ ,\ Y''\big|_{\omega\to0} = \infty\ \ ,\qquad Y''\big|_{\omega\to\infty} = 0$$

$$\varepsilon'\big|_{\omega\to0} = \left(\frac{C_{11}+C_{12}}{C_0}\right),\ \varepsilon'\big|_{\omega\to\infty} = \frac{C_{12}}{C_0}\ ,\qquad \varepsilon''\big|_{\omega\to0} = 0\ ,\ \varepsilon''\big|_{\omega\to\infty} = 0 \qquad\qquad \dots(8)$$

It is known that the above circuit shows two resonance frequencies given by $1/\sqrt{(L_{11}C_{11})}$ and $1/\sqrt{(L_{11}C_{11}C_{12}/(C_{11}+C_{12}))}$ for low R_{11} [18]. These so called resonance and anti resonance frequencies may be obtained by finding the frequencies where Z" equals zero. Generally these two frequencies are very close to each other. The variation of absolute impedance as function of frequency is shown in   Fig. 3.
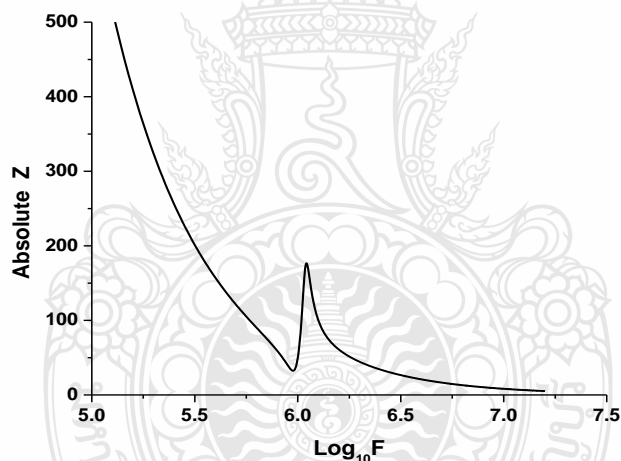

Fig 3. Plot of Absolute value of Z vs Log$_{10}$(F)

It is interesting to note that information about the circuit is contained within these two frequencies. Let us analyze the behaviour near the resonance frequency $\omega_r = 1/\sqrt{(L_{11}C_{11})}$. It can be shown that the values of Z' and Z" at frequency ω close to ω_r , say $\omega = \omega_r + \delta\omega$, are given by

$$Z' = \frac{R_{11}}{\left[1 - C_{12}\,L_{11}\,2\omega_r\delta\omega\right]^2 + \left[\omega\,C_{12}R_{11}\right]^2} \qquad\qquad \dots(9)$$

and

$$Z'' = \frac{1}{\omega_r\,C_{12}}\left\{\frac{-1 + \omega_r C_{12}\,L_{11}\,2\delta\omega}{\left[1 - C_{12}\,L_{11}\,2\omega_r\delta\omega\right]^2 + \left[\omega_r\,C_{12}R_{11}\right]^2}\right\} \qquad\qquad \dots(10)$$

It is found that Z' and Z" satisfy the relation

$$\left[Z'-\frac{1}{2}\left(\frac{1}{\omega_r C_{12}}\right)^2\frac{1}{R_{11}}\right]^2+\left[Z''-\frac{1}{\omega_r C_{12}}\right]^2=\left[\frac{1}{2}\left(\frac{1}{\omega_r C_{12}}\right)^2\frac{1}{R_{11}}\right]^2 \qquad \ldots(11)$$

This is the equation of a circle. Thus Z" vs. Z' plot would be a circle with centre at $(1/(2\omega_r C_{12} R_{12}), 1/(\omega_r C_{12}))$ and radius $1/(2\omega_r C_{12} R_{12})$, Similarly it is found that real and imaginary parts of Y, ε and M satisfy the following relations

$$\left(Y'-\frac{1}{2R_{11}}\right)^2+\left(Y''-\omega_r C_{12}\right)^2=\left(\frac{1}{2R_{11}}\right)^2 \qquad \ldots(12)$$

$$\left(\varepsilon'-\frac{C_{12}}{C_0}\right)^2+\left(\varepsilon''-\frac{1}{\omega_r C_0 R_{11}}\right)^2=\left(\frac{1}{2\omega_r C_0 R_{11}}\right)^2 \qquad \ldots(13)$$

$$\left(M'-\frac{C_0}{C_{12}}\right)^2+\left(M''-\frac{C_0}{2\omega_r C_{12}C_0 R_{11}C_{12}}\right)^2=\left(\frac{C_0}{2\omega_r C_{12}C_0 R_{11}}\right)^2 \qquad \ldots(14)$$

The simulated plots of the immittance functions are obtained and are not shown here. However they are similar to those shown in Fig. It can be seen that knowledge of C_0 and limiting values of ε' would yield estimates for C_{11} and C_{12}. Also complex plane plots of Y" vs. Y' measured at frequencies close to resonance would yield a circle with center as $(1/(2R_{11}), (\omega_r C_{12}))$ and radius at $1/(2R_{11})$. Values of L_{11} can be estimated from the resonance frequencies.

4. Calculation of correct values of R_{11}, L_{11}, C_{11} and C_{12}

Now we demonstrate that correct values of the equivalent circuit components R_{11}, L_{11}, C_{11}, and C_{12} can be obtained by fitting the experimental values of any of the immittance function to the corresponding expression by applying the method of Complex Nonlinear Least Squares (CNLS) [12,15, 19] and using the values obtained earlier by looking at the plots as initial guesses.

For this we generated immittance data by using the values of R_{11}, L_{11}, C_{11} and C_{12} as taken by Sherrit [6] viz. R_{11} = 37.67 Ω, L_{11} = 5.420 x 10^{-5} H, C_{11} = 4.987 x 10^{-10} F, C_{12} = 1.945 x 10^{-9} F and plotted as function of frequency as well as in complex plane. These are shown by dark circles. For the demonstration of our procedure we treat these immittance values as experimentally observed values. By looking at the experimental values of ε' vs. $\log_{10}$F as F tends to infinity and to zero we get C_{12}/C_0= 800, and $(C_{11}+C_{12})/C_0$ = 1300 from the low frequency and high frequency ends. Taking C_0 = 2x 10^{-12} F which is the value of the capacitance of the empty cell containing two parallel plates of diameter 10 mm and separated by 1 mm as is usually the case in our laboratory we find C_{11}= 4 x 10^{-10} F and C_{12}= 1.6 x 10^{-9} F. Now looking at the Y" vs. Y' plot which is a circle and at its centre as shown by the dark points in Fig.6 and using Eq. 12) the value of R_{11} is estimated to be 36.4 Ohm as obtained from the radius of the circle. As commented earlier Y" vs Y' plot is a circle with radius ($1/(2R_{11})$ and centre at ($1/(2R_{11})$, $(\omega_r C_{12})$) . Using the value of C_{12} obtained above and the Y coordinate of the centre from the graph which is equal to 0.0112 we get the value of ω_r as 7 x 10^6. This yields a value of 1.1x10^6 Hz as the resonating frequency. Now using the formula $\omega_r=1/\sqrt{(L_{11}C_{11})}$ the value of L_{11} is obtained as 5.1 X 10^{-5} H. These rough estimates ie R_{11} =36.4 Ω ,L_{11} =5.1 x 10^{-5} H , C_{11}= 4 x 10^{-10} F and C_{12}=1.6 x 10^{-9} F are used as initial guesses and their correct values are obtained by fitting the experimental data to the expressions of the immittance functions given above in Eqns. (11) to (14) [19]. The correct values are (37.670 ±1.169) x10^{-6} Ω, (5.420±0.001) x10^{-5} H, (4.987 ±0.001) x 10^{-10} F and (1.945 ±0.001)x 10^{-9} F respectively. The fitted and experimental values of Z*, Y*, M*, ε* are shown in Fig.4 where continuous line show the value of immittance function calculated after the fit. It is to be emphasized that this method can be widely used for determining the values of components used in other models of piezoelectrics as well [20].
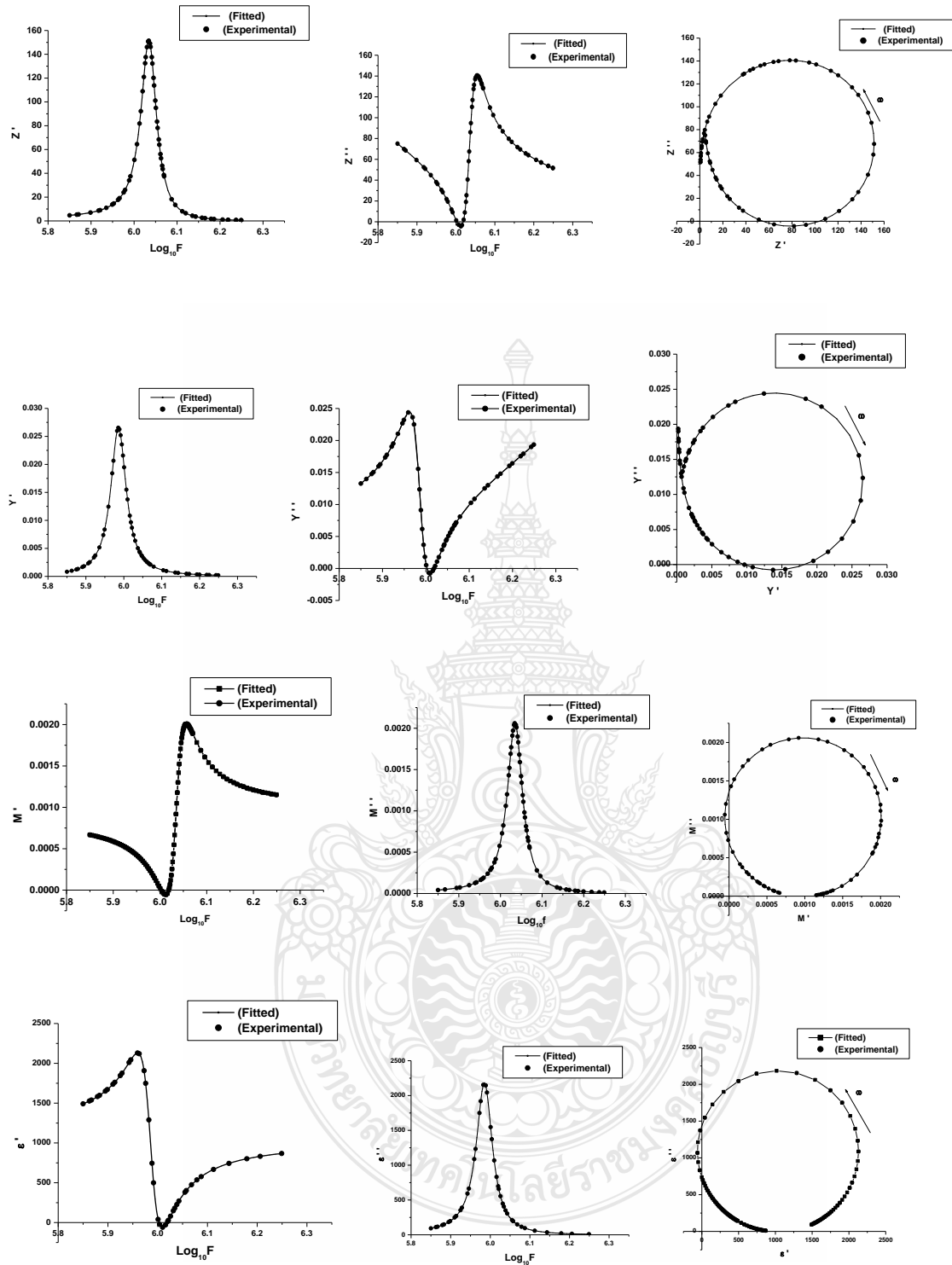
Fig.4 Experimental and fitted plots of real and imaginary parts of Z ,Y, M and ε vs.

Log$_{10}$(F) and their corresponding complex plane plots.

References

[1]   A. J. Moulson and J. M. Herbert, "Electro-Ceramics", Chapman and   Hall, New York, (1990).

[2]   R. B. Buck, E. Lindner, W. Kutner and G. Inzelt,   Pure. Appl. Chem., 76, 1139 (2004).

[3]   C. K. Ho, A. Robinson, D. R. Miller and M. J. Davis. Overview of Sensors and Needs for Environmental Monitoring. Sensors 2005, 5, 4-37

[4]   D. A Buttry and M. D. Ward, Chem. Rev., 92, 1355 (1992).

[5]   J. Singh, N. C. Soni and S.L. Shrivastava, Bull. Matter. Sci., 26(4), 397(2003).

[6]   S. Sherrit, H. D. Wiedarick, B. K. Mukherjee and M. Sayer, J. Phys. D: Appl. Phys., 30, 2354(1997).

[7]   E. Markiewicz, J. Kulek and Cz. Pawalacyzk, IEEE Trans. Dielectric and Electrical Insulation 6(3), 304(1999).

[8]   T. Tsurumi, T. Ichihara, K. Asaga and M. Daimon, J. Am. Ceram. Soc., 73(5), 1330 (1990).

[9]   J. S. Kim, K Choi, L. Yu., IEEE Trans. On ultrasonics, Ferroelectrics and frequency control, 40 (4), 424 (1993).

[10] W. G. Caddy, Piezoelectricity, Mc Graw Hill Book co., New York (1946).

[11] M. Aparna, T. Bhimasankaran, S.V. Suryanarayana, G. Prasad and G. S. Kumar, Bull. Mater. Sci., 24, 497 (2001).

[12] L. Pandey, O. Parkash, R.K. Katare and D. Kumar, Bull. Matter. Sci., 18, 563 (1995).

[13] F.C. Fonseca, J.A. Souza, R.F. Jardim, R. Muccillo, E.N.S. Muccillo, D. Gouvea, M. H. Jung and A. H. Lacerda, Phys. Stat. Sol. A, 199 255 (2003).

[14] F.C. Fonseca, J.A. Souza, R.F. JArdin, R. Muccillo, E.N.S. Muccillo, D. Gouvea, M. H. Jung and A. H. Lacerda, J. European Ceram. Soc., 24 1271 (2004).

[15] J. R. Macdonald, Impedance Spectroscopy, John Wiley and Sons, New York (1987).

[16] A. K. Jonscher, Dielectric Relaxation in solids, Chelsa Dielectric Press, London (1983).

[17] L. Pandey, R. K. Katare, O. Parkash, D. Kumar and O.P. Thakur, Ind. J. Pure & Appl. Phys., 36, 228(1998).

[18] R. B. Buck, E. Lindner, W. Kutner and G. Inzelt,   Pure. Appl. Chem., 76, 1139 (2004).

[19] L. Pandey, Workshop on use of computer in teaching Physics, partly sponsored by ICTP, Jabalpur, India (1992).

[20] Pramod Chaitanya, Impedance Spectroscopy and RF Pulse Response of Piezoelectric Materials, Ph.D. Thesis, Rani Durgavati University, Jabalpur, India (2008).