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Dielectric Behaviour of Pzt Ceramics at Microwave Frequencies

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ABSTRACT

The dielectric behavior of $PbZr_{0.52}Ti_{0.48}O_3$ have been carried out at the X-band microwave frequency. The composite material was prepared by using solid state reaction method. The dielectric constant (ε'), dielectric loss (ε"), quality factor ($Q = \frac{1}{tan\delta}$), relaxation time (τ) and conductivity (σ) of PZT having particle sizes 500, 250,176.5and125 microns have been studied at different temperatures i.e. -10^0,$+10^0$,$+30^0$ and $+50^0$c. The experimental dielectric values have been verified by using correlation formulae's of Landau-Lifshitz-Looyenga and Bottcher. It founds the good agreement with experimental values.

KEYWORDS: *Dielectric properties, pzt, ferroelectric ceramics*

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I. INTRODUCTION:

Perovskite $P_bZr_{1-x}Ti_xO_3$ ceramics are the most widely used Ferroelectric materials because & their excellent properties. The highest piezoelectric and electromechanical coupling co-efficient have been obtained for compositions nears the morpho tropic phase boundary (MPB) between rhombohedra and tetragonal phase. Piezoelectricity was first discovered by Jacques and Pierce Curie in 1880, and other materials later discovered included Rochelle salt, $BaTio_3$, P_bTio_3, ($P_bz_{R0.52}$ $Ti_{0.48}o_3$), etc. Since lead zircon ate titanate (PZT) her excellent piezoelectric properties, a high curies temperature, high spontaneous polarization and high electromechanical coupling co-efficient it has become an important industrial product.

The compound P_b ($Zr_{0.52}$ $Ti_{0.48}$)O_3 (PZT) has great piezoelectric properties which can apply in transducer applications [1]. The optimized PZT thin film was also to successfully fabricate an acoustic device. The device could be used as a transmitter or as a receiver [2]. PZT is one of the best know Ferroelectric material due to its remarkable ferroelectric and piezoelectric properties in polycrystalline from [3-5]. It is most widely used piezoelectric ceramic materials in devices like actuators, ultrasonic, transducers, sensors, resonators, ferroelectric memory, optoelectronic, piezoelectric transformers [6-8]. Study of dielectric materials in microelectronics devices gave rise to a new generation of microelectronic devices, such as microelectromechanical systems, non-volatile random access memory microwave integrated circuits [9]. The dielectric, elastic and piezoelectric properties of Ferroelectric materials are dependent on frequency and amplitude of the driving field. Materials exhibiting piezoelectricity as well as nonlinear dielectric behavior are named as Ferroelectric materials. These types of materials have wide range of applications as sound wave detector, phonograph, pickups, microphones, accelerometers, biomedical field, capacitors etc.[10]. The main objective of this work is to prepare the PZT material and to study its dielectric parameters for various grain sizes and temperatures. The similar work has been done by other researchers [11-14].

II. EXPERIMENTAL PROCEDURE:

Synthesis of material : PZT composite material was prepared for microwave studies by using solid state reaction method. In SSR method, the solid reactants react chemically without presence of any solvent at high temperatures yielding a product which is stable. The advantage of SSR method is that final product in solid form is structurally pure with the desired properties depending on the final sintering temperatures. This method is environment friendly and no toxic an unwanted waste is produced after the SSR is complete. The steps involved in SSR are –

[1] Take weighed appropriate high purity starting materials, fine grain powders in stoichiometric properties.
[2] Mix them together, thoroughly using agate mortar and pestle or ball milling.
[3] Heat the solid powder mixture (calcinations) at elevated temperatures in air using muffle furnace.
[4] Report the calcinations process twice with intermittent grinding.
[5] Final powder is ready.

The high purity AR grade Pb_3O_4 $ZrO_2T_iO_2$ were used as starting materials for the solid state reaction. [15]. These constituents were weighed and mixed thoroughly. The composite material was prepared for 50gm. Pb_3O_4 ZrO_2 & T_iO_2 are estimated as per P_b $Zr_{0.52}Ti_{0.48}$ O_3 so that total molecular wt.is 318.424gm. Then after estimating the samples of weights Pb_3O_4 ZrO_2 & T_iO_2 had taken as 212.536, 73.932 and 31.956 gms respectively out of 50 gms. Then this was mixed in stoichiometric proportion has been grinded about 3½ hours continuously using agate mortar for first calcinations. This mixture was initially sintered at temp 800^0c for a time about 4 hours in a muffle furnace. Then after first calcinations the sample was again grinded or 1½ hrs and kept about 4 hrs. at 1200^0c for final sintering. The chemical reaction at high temperature take place, through solid state reaction giving a stable sample product free of residual reactants.

Material preparation for Dielectric studies : For the determination of dielectric parameters of PZT were prepared by using sieves of different sizes. Like 500, 250, 176.5, 125 microns particle sizes. All the samples transferred into the glass bottles and labeled according to their particle size. To determine the relative packing factor (δr) densities for each powder sample is measured, measurement of dielectric parameters (ε′) and (ε″) for these powder samples were carried out using reflectometer technique at 9.85 GHz [10 to 13] microwave frequency and at temperatures (-10^0c, $+10^0$c, $+30^0$c & $+50^0$c).For the accurate measurement of wavelength in dielectric (λ_d), sample is introduced in the dielectric cell in steps. Applying constant force of 98N on the sample, and for each time the corresponding output power is measured by using crystal pick in the directional coupler. The relationship between reflected power and sample height is approximately given by a sinusoidal curve. The distance between two adjacent minima of the curve gives half the dielectric wavelength (λ_d).

Determination of molecular parameters: The dielectric constant (ε′) and loss factor (ε″) for PZT powder at microwave frequency are determined by using relations [16].

$$\epsilon_p^{'} = \left(\frac{\lambda_0}{\lambda_c}\right)^2 + \left(\frac{\lambda_0}{\lambda_d}\right)^2 \qquad .... (1)$$

$$\epsilon_p^{"} = \frac{2}{\pi}\left(\frac{\lambda_0}{\lambda_c}\right)^2 \frac{\lambda_g}{\lambda_d}\left(\frac{d_{pmean}}{d_n}\right)^2 \qquad .... (2)$$

Where λ_0 is the free space wavelength

λ_d is the wavelength in dielectric

λ_c is the cutoff wavelength of waveguide

λ_g is the guide wavelength

The conductivity (σ_p) and relaxation time (τ_p) are obtained by using following relations.

$$\sigma_p = \omega\epsilon_0\epsilon^{"} \qquad ......(3)$$

$$\tau_p = \epsilon^{"}/\omega\epsilon^{'} \qquad ......(4)$$

Where w = 9.85 GHz angular frequency of measurement ε_0 is the permittivity of free space.

The values of $\epsilon_s^{'}$ and $\epsilon_s^{"}$ for bulk materials can be co-related by using the relations given by Bottcher and Landau – Lifshitz – Looyenga.

III. RESULTS AND DISCUSSION :

Table 1 shows values of permittivity ($\epsilon_p^{'}$) and loss factor ($\epsilon_p^{"}$) along with values of relative packing fraction (δr) for different particle sizes and temperatures. There is systematic increase in $\epsilon_p^{'}$ and $\epsilon_p^{"}$ with increasing δr. and systematic decrease in $\epsilon_p^{'}$ and $\epsilon_p^{"}$ with increasing temperature. This is expected because at higher values of δr the interparticle hindrance offered to the dipolar motion for a compact medium is much higher than less bounded particles. The calculations of quality factor (Qxf) where f is resonant microwave frequency are also mentioned in table 1. It is found decrease in Q x f values for decreasing particle size and by increasing temperature the Q x f values increases. The values of relaxation time (τ_p) and conductivity (σ_p) are increases systematically by increasing δr and decreases by increasing of temp. It is due to, when polar molecules are very large, then under the influence of high frequency the rotary motion of polar molecules of a system is not sufficiently rapid to attain equilibrium with field. The increase in τ_p by increasing δ_r is due to increasing hindrance to the process of polarization. The increase in σ_p suggests that at higher compaction, no micro cracks develop in sample due to high mechanical pressure. As temp increases, τ_p decreases may be due to increase in the effective length of dipole. Again increase in temp, causes an increase in energy loss due to the large number of collisions and thereby decreasing τ_p.

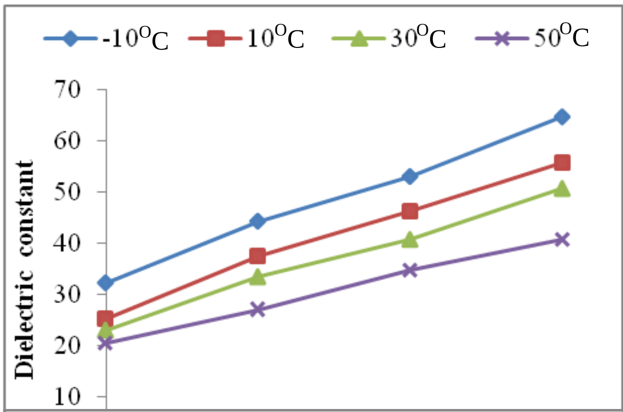
Table 2 indicates the measured and computed values of $\epsilon_s^{'}$ and $\epsilon_s^{"}$ for bulk from powder measurements. The results reported at δr = 1 are those measured on the finest crushed powder sample packed at very closely in a sample holding dielectric cell at 98N force, so minimum voids between the particles. The smallest particle size 125 micron or less assumed this system as solid bulk for getting correlation between powder and solid bulk. The correlation formulae of Landau – Lifshitz – Looyenga and Bottcher was used. The bulk values $\epsilon_s^{'}$ and $\epsilon_s^{''}$ are very much closer to measured values.

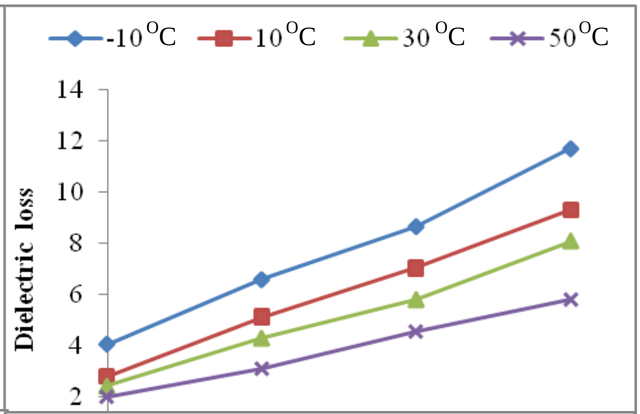
Table 1. Values of $\epsilon_p^{'}$, $\epsilon_p^{''}$, Q x f, τ_p and σ_p of PZT at different temperatures and particle sizes.

Temperature (°c)	Relative packing factor (δr)	$\epsilon_p^{'}$	$\epsilon_p^{''}$	$Q=1/tan$	Q x F GHz	τ_p (P.S.)	σ_p
-10	0.86	32.35	4.08	7.93	78.11	2.04	2.23
	0.90	44.41	6.60	6.73	66.33	2.40	3.61
	0.94	53.18	8.67	6.13	60.43	2.64	4.71
	1.00	64.87	11.71	5.54	54.60	2.92	6.41
10	0.86	25.45	2.83	8.99	88.58	1.78	1.55
	0.90	37.66	5.14	7.33	72.16	2.21	2.81
	0.94	46.38	7.05	6.58	64.80	2.45	3.85
	1.00	55.78	9.32	5.98	58.95	2.71	5.10
30	0.86	23.16	2.45	9.45	93.10	1.71	1.34
	0.90	33.56	4.31	7.78	76.65	2.07	2.36
	0.94	40.82	5.81	7.03	69.22	2.30	3.17
	1.00	50.76	8.08	6.28	61.87	2.57	4.42
50	0.86	20.56	2.04	10.07	99.19	1.60	1.11
	0.90	27.17	3.13	8.68	85.58	1.86	1.71
	0.94	34.85	4.57	7.63	75.13	2.11	2.50
	1.00	40.82	5.81	7.03	69.22	2.30	3.17

Table 2. Measured and calculated Values of $\epsilon_s^{'}$ and $\epsilon_s^{''}$, for solid bulk from PZT at different temperatures and packing fractions.

Temperature (°c)	Relative packing factor (δr)	Measured value $\epsilon_p^{'}$	Calculated from Bottcher's $\epsilon_s^{'}$	Calculated from L-L-L $\epsilon_s^{'}$	Measured value $\epsilon_p^{''}$	Calculated from Bottcher's $\epsilon_s^{'}$	Calculated from L-L-L $\epsilon_s^{'}$
- 10	0.86	32.35	40.52	39.74	4.08	5.51	5.16
	0.90	44.41	51.19	50.68	6.60	8.14	7.76
	0.94	53.18	58.28	56.66	8.67	9.80	9.52
	1.00	64.87	64.87	64.87	11.71	11.71	11.71
+ 10	0.86	25.45	31.79	31.78	2.83	3.81	3.58
	0.90	37.66	44.03	44.02	5.14	6.33	6.04
	0.94	46.38	50.82	50.81	7.05	7.97	7.74
	1.00	55.78	55.78	55.78	9.32	9.32	9.32
+ 30	0.86	23.16	28.89	28.50	2.45	3.30	3.1
	0.90	33.56	39.20	38.48	4.31	5.31	5.07
	0.94	40.82	44.70	43.75	5.81	6.56	6.38
	1.00	50.76	50.76	49.14	8.08	8.08	8.08
+ 50	0.86	20.56	25.60	25.30	2.04	2.74	2.58
	0.90	27.17	31.69	31.22	3.13	3.85	3.68
	0.94	34.85	38.14	37.22	4.57	5.16	5.02
	1.00	40.82	40.82	40.03	5.81	5.81	5.81





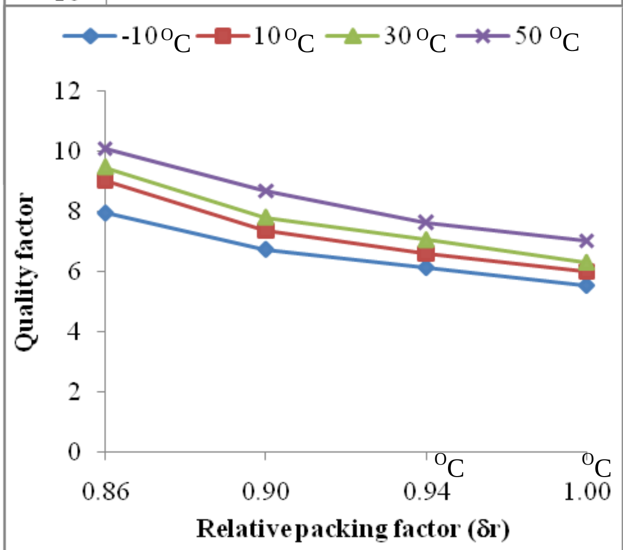


Fig.3 δr Vs Q

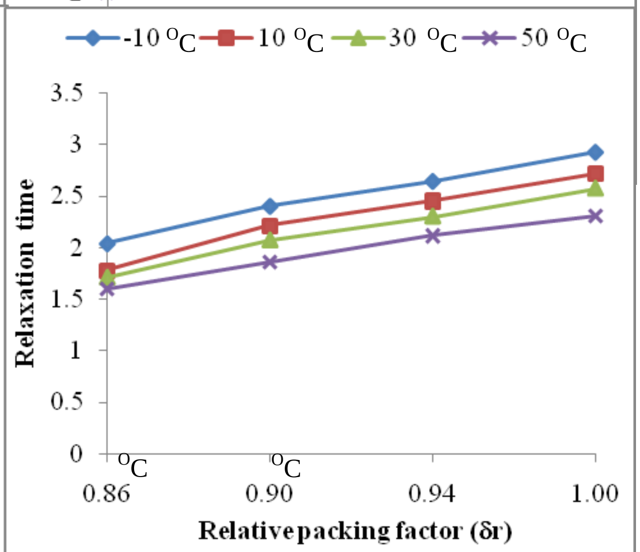


Fig.4 δr Vs τ

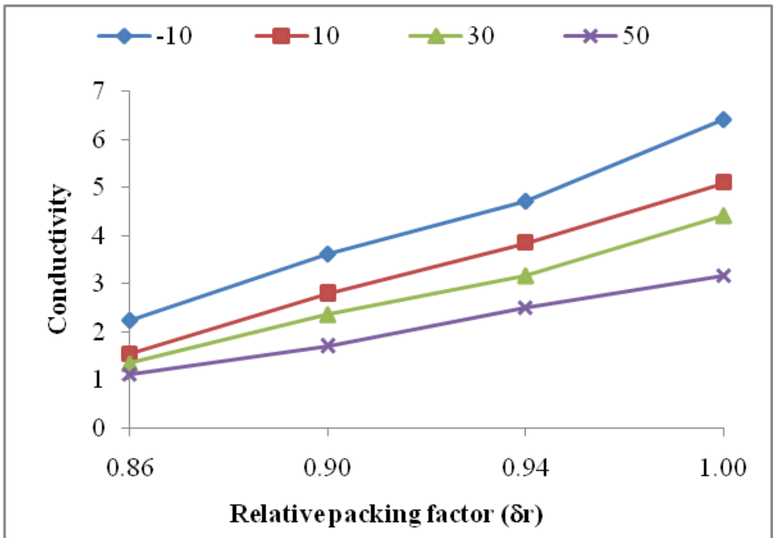


Fig.5 δr Vs σ

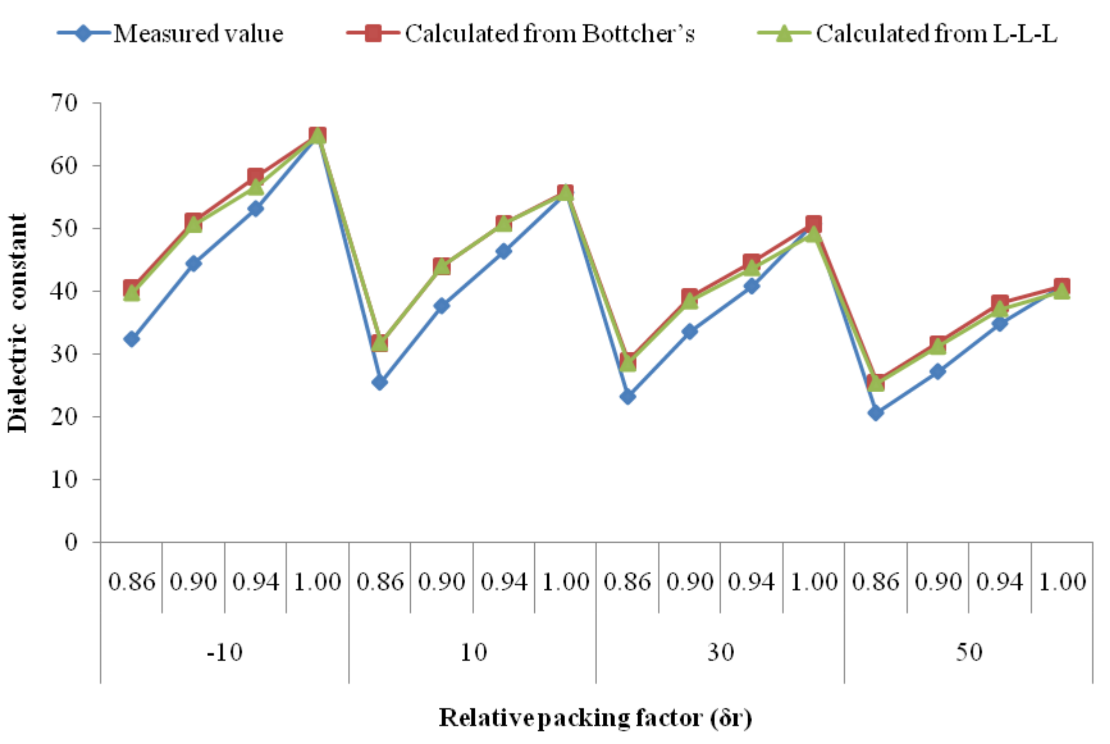


Fig.6. δr Vs ε' (Exp, Bott., L.L.L)

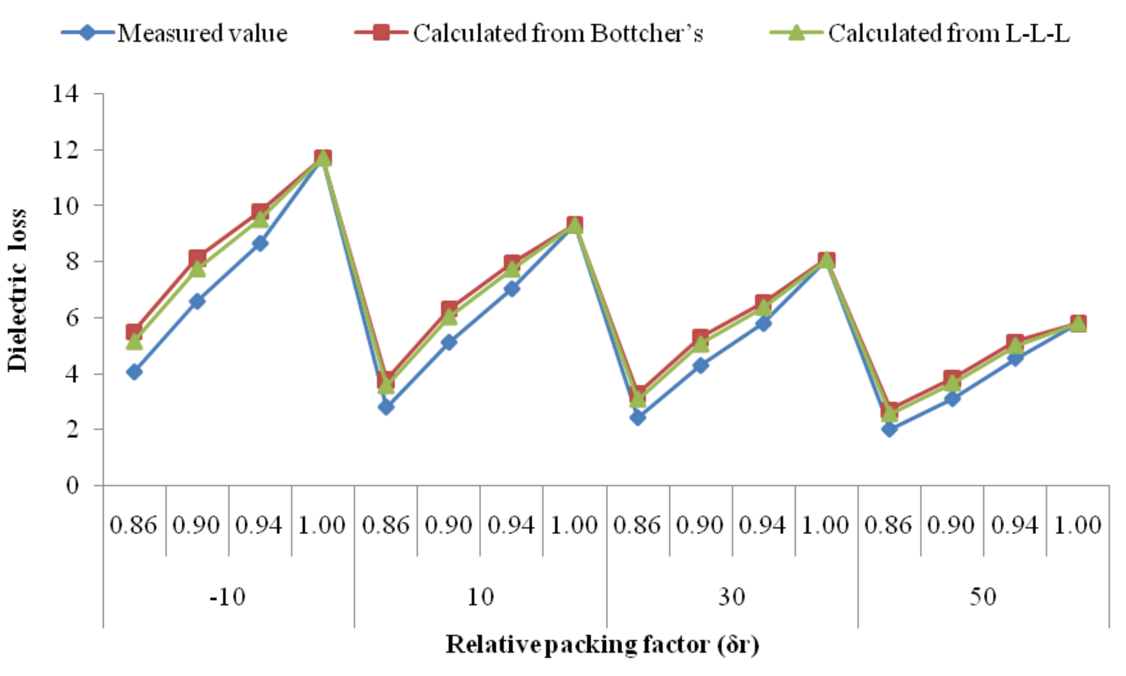


Fig.7. δr Vs ε'' (Exp, Bott., L.L.L)

IV. CONCLUSION:

It is found there is fair agreement of values dielectric constant between values obtained experimentally and theoretically given by Landau – Litshitz – Looyenga and Bottcher between powder and solid bulk works well.

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REFERENCES

[1] Wanwilai Chaisan *et al.,* Dielectric and ferrolectric properties of lead zirconate titanate caramics prepared by modified mixed oxide method, *Material Chemistry and Physics*, 104, 2007, 113-118.
[2] Chung-Cheng Chang *et al.,* The Farbication and characterization of PZT Thin film Acoustic devices for Application in underwater Rototic system. *Proc. Natl. Sci. Counc. Roc.(A),* 24(4), 2000, 287-292.
[3] Y. Xu, Ferroelectric materials and their applications, (North Holland, Amsterdon, 1991).
[4] G. Saghi-Szabo and R.E. Cohen, First principles study of piezoelectricity in tetragonal $P_bT_iO_3$ and $P_bZ_{r1/2}T_{i1/2}O_3$. *Phys. Rev.* A, 59(20)
[5] Z. Zhang, P. We, K.P. Ong, L. Lu and C. Shu, Electronic properties of A-site substituted lead zirconate fitanate; density functional calculations, *Phys. Rev.* B, 76 (125102), 2007.
[6] R.C. Buchanan, Ceramic materials for electronics, (Marcel Dekker, New York, 1991).
[7] G.H. Haexptling, Ferroelectric ceramics : History and technology, *J. Am. Ceram. Soc.,* 82, 1999, 797.
[8] P. Muralt, J. Micromech, Microeng, 10, 2000, 136.
[9] J.F. Scott, Ferroelectric Review, 1, 1998, 1-129.
[10] P.G Gawali and B.K Bongane, Dielectric behavior & some pulses using reflectmetric technique at 9.85 CH_3, *Indian Journal & applies research,* 3, 2003.
[11] B.S Narawade, P.G Gawali & G.M Kalmse, Dielectric studies & Binary mixture of n-propyl alcohol ethylenediamine, *Journal of Chemical Sciences & India Academy of Sciences,* 117(6), 2005, 673-677.
[12] P.G. Gawali and B.K Bongane, Microwave dielectric properties of $(Ba_{o.5}Pb_{o.5})Tio_3$ with wave various temperature and particle sizes, *Bio-nano Frontier (Communicated)*.
[13] B.K Bongane and P.G Gawali. Effect of particle size and temperature on dielectric parameters of PLZT ceramics at 9.85 GH_3, *International Journal of Physics and Mathematical Sciences* (Accepted MS NO. IJPMS 2014/04/011).
[14] N.S Gajbhiye *et al.*, Dielectric properties of nanostructure PZT synthesized by chemical methods, Defense Science Journal, 57(1), 2007, 61-68.
[15] Mridula Kumari, Arun Singh, Jagdhar Mandal, Structural and Dielectric properties of PZT ceramics prepared by solid state reaction route. *International Journal of Scientific & Engineering Research,* 5(4) 2014, 404-406.
[16] Yadav J.S. and Gandhi J.M., Simple microwave technique for measuring the dielectric parameters and their powders, *Indian Journal of Pure and Applied Physics,* 30, 1992, 427.