Electrical Characterization of PZT Dielectric Thick Film with varying Zr/Ti ratios Developed by SOL GEL Technique.

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Abstract - After being reported about various parametric changes in piezoelectric coefficient, polarization and dielectric constant K by addition of some foreign ions; a new dimension was added in classifying PZT composite material as hard and soft doped. As reported by various researchers across the globe, soft PZTs have highly enhanced piezoelectric properties which makes them highly suitable for fabrication of various electrical and electronic devices owing to high gain and better response, but on the other hand hard PZTs are reported to have highly decreased dielectric constants and piezoelectric coefficients leading their incapability to serve as an effective material for fabrication of actuators. However a considerable linearity in the variation of K is reported in these hard doped materials, which can indirectly be utilized for other low gain and low response systems. In the presented work, the author investigates the nature of variation in Curie - lattice parameters and dielectric constant composite material Pb[Zrx,Ti(1-x)]O3, for x=0.6, 0.7and 0.8; furthermore the effect of hard dopant Mg+2, on the composite material is investigated providing mathematical results which are in coherence with the pre established theory.

Keywords*: PZT, nanoceramics.*

1. Synthesis of $Pb[Zr_XTi_{1-x}]O_3$ Nanoceramics using Sol-Gel Technology

The synthesis of $Pb[Zr_XTi_{(1-x)}]O_3$ nanocramics is carried out by Sol-Gel process with the raw materials lead acetate trihydrate trihydrate $Pb(CH_3COO)_2.3H_2O$, zirconium propoxide $Zr(C_3H_7O)_4$ and titanium iso-propoxide $Ti[(CH_3)_2CHO]_4$.

To start with, lead acetate is dissolved in acetic acid in proper stochiometric ratios for different values of x, (x=0.6, 07 and 0.8) and is heated at 110°C for an hour to remove water and then cooled down to 70oC. With constant stirring using magnetic stirrer, zirconium propoxide followed by titanium isopropoxide is added in a calculated ration corresponding to x=0.6, 0.7 and 0.8 respectively, to the above mixture. Ethylene glycol is added to the above mixture in the ratio of 1 ml of glycol to 10 gm of lead acetate. The initial reaction had to be completed before glycol is added since residual zirconium propoxide and titanium isopropoxide alcolyze with glycol to form a condense solid [1]. A small amount of distilled water is added to get the final sol. The sol is kept at 70°C for 24 h to get the clear transparent gel. The gel is then dried in a controlled oven at 100°C for 48 h to get a light brown powder. The oven dried gel is calcined at 550°C for 5 h. After proper grinding the sample powders corresponding to different values of x that is Pb[Zr0.6Ti0.4]O3, Pb[Zr0.7Ti0.3]O3 and Pb[Zr0.8Ti0.2]O3 are each divided into four parts by weight. To three parts 3, 5, 7 weight % MgO was added followed by proper mixing. The samples were cold pressed at a pressure of 6x107 kg/m2 using a hydraulic press to form disks shaped pallets 1–2 mm thick. These pallets were calcined at 700°C for 5 h, followed by sintering at 1300°C for 2 h. PbZrO3 powder was used as a setter during sintering in order to prevent PbO loss or vaporization at higher temperatures. Density of each sample was calculated by dimensional measurement of thickness and diameter using precision Instruments. Pb(CH3COO)2.3H2O, zirconium propoxide Zr(C3H7O)4 and titanium iso-propoxide Ti[(CH3)2CHO]4. To start with, lead acetate is dissolved in acetic acid in proper stochiometric ratios for different values of x, (x=0.6, 07 and 0.8) and is heated These pallets were calcined at 700°C for 5 h, followed by sintering at 1300°C for 2 h. PbZrO3 powder was used as a setter during sintering in order to prevent PbO loss or vaporization at higher temperatures. Density of each sample was calculated by dimensional measurement of each using high precision instruments in lab.

2. EXPERIMENTAL RESULTS AND DISCUSSIONS

Determination of Lattice Parameters

Lattice parameter of each developed sample of $Pb[Zr_xTi_{(1-x)}]O_3$ corresponding to x=0.6, 0.7 and 0.8 are determined by well-known Scherrer's formula , thereby studying the nature of Curie temperature variation with the variation in grain size (N). Interestingly, it is noted by the author after studying the nature of variation, that the Curie temperature falls significantly, however the nature of fall is slow for N>15, but is drastic for N<15, where N is the number of layers. The lattice parameter, is approximately found to be 0.4 nm which is in accordance with the value reported by Glazer *et al.* [2]. Therefore the grain size for N=15 is about 12 nm. Further calculations confirm the nature to be slight irregularity in the variation for sample the 80/20 composition near N=10 to 12 and highly nonlinear for N=30 and above. Therefore the composition PZT(80/20) is considered as unsuitable by the author for the fabrication. [3]

Study and Analysis of the Variation of Dielectric Constant with N (Grain Size)

Variation of Dielectric Constant K with grain size for different compositions is studied and plotted in Figure 1. This curve confirms that the choice of the material composition as mentioned above by the author is appropriate out of the three compositions corresponding to x=0.6, 0.7 and 0.8 chosen.

As observed from the curve, the value of the dielectric constant from N=3 to 6 gives anomalously high value for the composition corresponding to x = 0.6, whereas in comparison though the value of dielectric
constant value is very high for x = 0.7, (approximately 2000), still remains within the measurable range. It has been established by various authors that $Pb[Zr_xTi_{(1-x)}]O_3$ has a non- linear variation of dielectric constant with temperature [4], which further increases by addition of soft dopants. This enhanced non linearity in dielectric constant is not desirous for designing of temperature sensors, hence author in this paper investigates the effect of hard doping of Mg^{+2} on the chosen sample corresponding to x=0.7, that is $Pb[Zr_{0.7},Ti_{0.3}]O_3$. The sample $Pb\ Zr_{0.7},Ti_{0.3}]O_3$ is variably doped with 3, 5 and 7 weight % Mg^{+2} and the results obtained is shown in Figure 3. It is observed that with increase in percentage doping of Mg^{+2}, the value of dielectric constant K falls significantly when compared with the undoped sample, with a significant increase in linearity [5]. At 7% Mg^{+2} doping; the material gives linear variation over a wide range of temperature, and interestingly the room temperature dielectric constant variation follows the same nature as the variation of density of the sample as shown in Figure 3. [6]

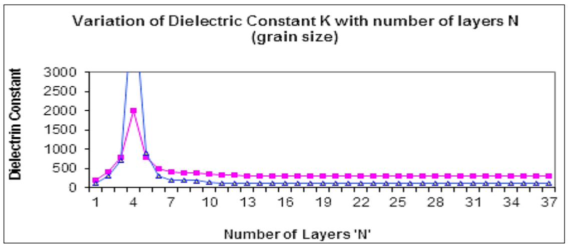

Figure -1: Variation of Dielectric Constant K with N (Pink and Blue Corresponding to the Composition 60/40 and 70/30 Respectively.

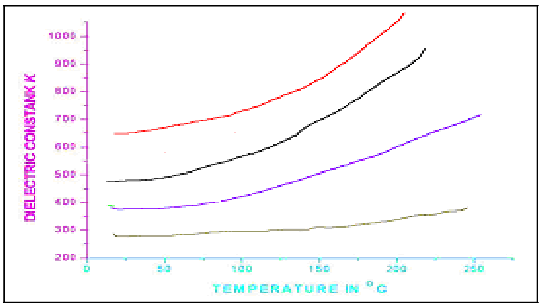

Fig -2: Variation of Dielectric Constant K with Temperature (Red, Black, Violet and Brown Curves Corresponds to the Undoped, 3%, 5%, 7% Doped Samples Respectively).

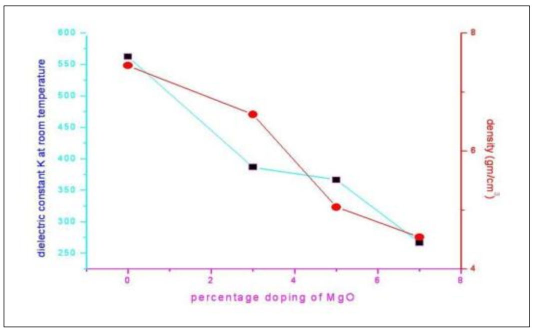

Fig -3: The Green and Red Curves Corresponds to Variation of K and Density in gm/cc Respectively with Percentage Doping.

3. CONCLUSION

It is finally concluded that all the samples of $Pb[Zr_x,Ti_{(1-x)}]O_3$ for x = 0.6, 0.7 and 0.8 synthesized by Sol-Gel process are homogeneous and formed in single rhombohedral phase. The sample corresponding to x=0.7 is highly compact, dense. After comprehensive studies and results obtained from the experiments it is confirmed that the composition corresponding to x=0.7, $Pb[Zr_{0.7}Ti_{0.3}]O_3$ is best suited as base material for the high dielectric constant makes it suitable for dielectric capacitor applications. Variable Mg^{+2} doping decreases the dielectric constant values considerably. Also it is evident that though the dielectric constant decreases significantly on increasing the doping percentage of Mg^{+2} there is significant improvement of linearity in the variation of dielectric constant with temperature, this is of great significance in designing thermal sensors as the complexity involved in the electronic circuitry to compensate for the non linearity can be reduced to significant level. Out of all samples 7% Mg^{+2} doped PZT is found to be best suitable and optimum among all the samples developed.

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