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RESEARCH ARTICLE

Study of Dielectric behavior and dielectric loss of PbZrO₃: Temperature dependence

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Abstract

The Perovskites PbZrO₃ is widely used in communication electronics, microelectronics, and anti-ferroelectric material. The dielectric behavior of the Perovskites PbZrO₃ is compressed in pellet form have been studied using capacity measuring unit Zeinith M92 A were synthesized by solid state reaction technique. The dielectric constant (ϵ') and dielectric loss (ϵ'') of the compounds have been studied at 1 kHz, 10 kHz and 100 kHz in the temperature range of 300K to 1200K. The dielectric constant of PbZrO₃ show rapidly increases above 700K and 850K respectively. After this temperature the dielectric constant decreases. The dielectric loss also shows similar behaviour as dielectric constant and it is also temperature dependent.

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INTRODUCTION

Dielectric materials are widely used in wireless communication devices. These require a high dielectric constant and low dielectric loss [1]. Lead Zirconate (PbZrO₃) are ABO₃ perovskite dielectric material that is both chemically and mechanically stable. They have unique dielectric properties that make them suitable for digital actuators and high charge storage capacitors [2]. The investigations reporting the dielectric properties of pure barium zirconate are 40-50 years old [3-5]. Hence, there are definite information gaps in the reported research in terms of the inevitable synthesis processing-microstructure property correlations

In this paper, we have discuss about the synthesis characterization and dielectric properties of PbZrO₃

Experimental:

Polycrystalline samples of PbZrO₃ compounds were synthesized by the solid state reaction technique, using high purity (99.99% pure) powders of PbO and ZrO₂, procured from Alfa Aesar, A Johnson Matthey, Chemical India Pvt. Ltd. The stoichiometric mixture of these oxides were mixed and heated in alumina crucible at temperature 1300K for 50 hrs. In this process the mixture was followed by one intermediate grinding and the final product was cool down slowly.

The X-ray diffraction of the compound was studied using x-ray diffractometer (Thermo electron-XRL EXTRA) at room temperature by using CuK α radiation with $\lambda=0.15418\text{nm}$ in a wide range of Bragg angle ($10^\circ \leq 2\theta \leq 90^\circ$). DTA, TGA and DTG studies of the compounds were carried out in nitrogen gas using a thermal analyzer (PERKIN ELEMER PYRIS) at a heating rate of 283K/min and flow rate of 100ml/min from 300 K to 1000 K.

The compounds were pressed at a pressure of $6.19 \times 10^8 \text{ Nm}^{-2}$ to form pellets of circular cross section (area $\sim 0.90 \times 10^{-4} \text{ m}^2$ and thickness $\sim 0.30 \times 10^{-2} \text{ m}$). The pellets were then sintered in air for 28hrs at 1500K. The pellet covered with film of silver paint on the opposite surfaces to obtain a good contact was inserted between the two silver electrodes. A LCR-Q meter (model-928, Systronic, India) was used to measure the capacitance(C) and quality factor (Q) of the sample at different temperatures and at a frequency of 1kHz. The dielectric constant (ϵ') and dielectric loss (ϵ'') of the sample was calculated by using the following relations [12-13]

$$\epsilon' = Cd / \epsilon_0 A$$

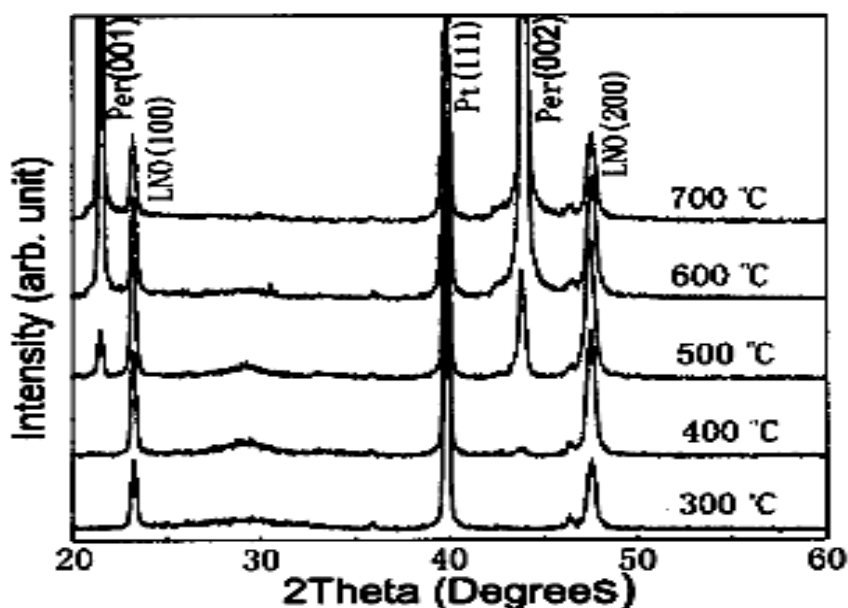
$$\epsilon'' = \epsilon' / Q$$

where C = the capacitance of the capacitor in Farad, d = thickness, A = face area of the pellet, ϵ_0 = the permittivity of free space and Q = quality factor respectively.

RESULT AND DISCUSSION

The XRD pattern of BaZrO₃ is shown in Figs. (1). From XRD pattern, d_{hkl} planes have been evaluated using relation [14].

$$d_{hkl} = \frac{0.15418}{2 \sin \theta}$$



ual procedure. All the peaks
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o), density of pressed

Compound	Unit cell	Lattice parameter			Density	Pore fraction
		^a 0	^b 0	^c 0		
		(nm)	(nm)	(nm)	(kgm ⁻³ x10 ⁻³)	
PbZrO ₃	Orthorhombic	0.68262	1.04412	0.58922	4.38	4.25
						0.028

The weight loss completed at 748K and 673K in PbZrO₃. Such a small loss in weight is due to removal of absorbed water and other gaseous species. No further weight loss appears over higher temperature which indicates that the compounds are stable and do not show decomposition up to 1123.

The DTG trace of PbZrO₃ a show maximum rate of mass change at 608K and 538K respectively The variation of dielectric constant (ϵ') and dielectric loss (ϵ'') and quality factor (Q) with temperature at 1 kHz is shown in Figs (2).

It is seen that these compounds have dielectric constant 25 and 39 for PbZrO₃ at 400K respectively. Since dielectric constant (ϵ') seems to have almost no temperature dependence, these values may be taken as the room temperature value of the materials. The reported value of ϵ' has been calculated using the capacitance of the pressed pellets. The density of these pellets remains less than theoretical density. This means pellet contains air pores. Therefore a correction for pore fraction (f_p) is essential to obtain the bulk value of dielectric constant (ϵ') and is given by [15].

$$f_p = \frac{d_o - d_p}{d_o}$$

For low conducting solids ϵ' and f_p are related by the relation

$$\epsilon_b = \frac{(\epsilon - f_p)^3}{1 - f_p}$$

The evaluated value of are 25 and 288 for PbZrO_3 . The value of becomes large as temperature is increased and validity of eq. (5) becomes doubtful. Further this formula changes only the magnitude of but not the nature of variation of with temperature. Therefore we have not used this correction at higher temperature.

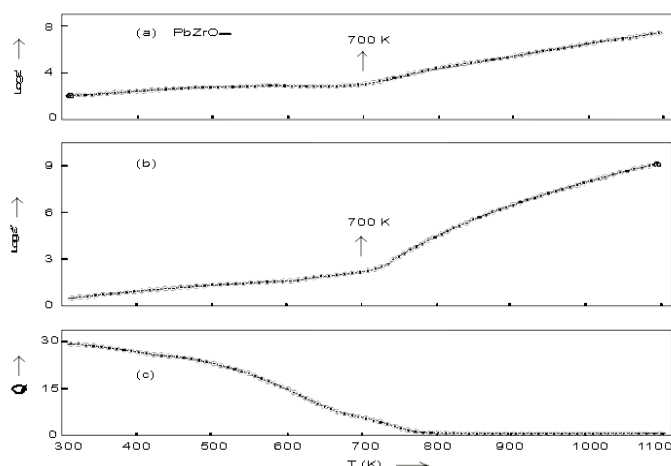
The values of dielectric constant (ϵ') and dielectric loss (ϵ'') of these compounds at different temperature and at frequency 1kHz are given in table 1 and 2 respectively.

Table-1 The dielectric constant for studied Zirconate at different temperature				
Compound	400K	Dielectric constant		
		600K	800K	1000K
PbZrO_3	25	39	61	1.12×10^3

Table-2 The dielectric loss for studied Zirconate at different temperature				
Compound	400K	Dielectric loss		
		600K	800K	1000K
PbZrO_3	2	3	1.58×10^1	2.00×10^3

It is seen that the systematic trend of and variation reveals that polarization mechanism in all these Zirconate is same centers and has larger polarizability. The dielectric constant has very slow increase at lower temperature. This shows that there is no chance for the existence of thermally generated charge carriers at lower side of temperature. The lower value of below 600K indicates that free charge carriers generated from impurities is also small. Well made electrode rules out the chance of interfacial polarization. Therefore this slow increase seems to be the combined effect of lattice and electronic polarizabilities of individual ions. The increase of these polarizabilities seems to compensate the slight decrease of polarizability due to decrease in the number of ions per unit volume following the lattice expansion with temperature. However, it must be noticed that the increase of with T is very small in comparison to the variation one expects for ionic solids. This indicates that either thermal expansion of these compounds is very small or they have some other kind of polarization mechanism.

The dielectric constant (ϵ') of these compounds have much faster increase above certain critical temperature (T_k). The dielectric loss (ϵ'') shows similar behavior above T_k . The critical temperature (T_k) is 820K and 700K for PbZrO_3 . The faster increase in dielectric constant (ϵ') above T_k is due to space charge polarization [17,18]. The pressed sample develops a considerable amount of space charge polarization arising out from the defects or impurities present in the bulk or at the surface of the material.



CONCLUSION

The XRD studies confirm that the studied compounds have single phase orthorhombic structure at room temperature. The dielectric constant and loss have very slow increase upto T_k . above T_k this increase becomes much faster. The value of T_k is different for both compounds. The reason for faster increase of ϵ' and ϵ'' above T_k is due to space charge effect of thermally generated charge carriers.

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