Observation of dielectric relaxor behavior in Pb$_{0.95}$ Sr$_{0.05}$(Zr$_{0.5}$Ti$_{0.5}$)O$_3$ ceramics

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ABSTRACT

We reports structural, microstructural and dielectric characteristics of Sr^{2+} doped PZT (50/50) ceramic. X-ray diffraction reveals that the system exhibit coexistence of two phases (Tetragonal and rhombohedral) at room temperature. Typical relaxor behavior was observed by the dielectric studies and confirmed by Vogul-Fulcher fitting. The observed relaxor was predicted as existence of nanopolar regions due to short range ordering in presence of oxygen vacancies. The evidence for oxygen vacancies was studied by conductivity and polarization studies. Moreover, at 270 °C one more phase transition is noted which was ascribed to structural phase transition. Present study has scientific significance to distinguish the performance of oxygen vacancies in ferroelectric materials. Copyright © 2015 VBRI press.

Keywords: Phase transition; hysteresis; oxygen vacancy; conductivity.

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Introduction

Lead zirconate titanate Pb(Zr$_{1-x}$Ti$_x$)O$_3$ has been extensively investigated in the form of single crystal, bulk as well as thin films, owing to (a) high piezoelectric response (b) high remnant polarization, (c) low coercive field (E$_c$) and (d) high Curie temperature (T$_c$) near to the morphotropic phase boundary, which are very much important for piezoelectric transformers and FeRAM applications and highly applicable for nonvolatile memory devices [1-4]. The reversible polarization can arise mainly from the ionic

displacement or displacement of the charged defects upon the application of the electric field. Whenever the field is removed, the polarization is relaxed and hence reversible polarization of ferroelectric material cannot be used for data storage. From the application point of view, the irreversible part of the polarization can be utilized in designing the memory devices [5, 6].

There have been several studies reported on divalent and trivalent cation doped PZT [7-11]. In these materials, morphotropic phase boundary (MPB) evolves due to the presence of both tetragonal phase and rhombohedral one. Connection of MPB with domain walls has already been explained earlier [12].The presence of rhombohedral phase increases the number of possible orientation of polarization. The domain wall energy calculation shows that (T-T twins T- tetragonal) exhibits 10^7 times higher elastic energy stored than (R-R twin R- rhombohedral). The formation of T-R twin at MPB causes a higher stability of ferroelectric domains which require a smaller energy for the rotation of polarization vector [12]. Therefore, domain wall movement plays a key role in the macroscopic response of ferroelectrics, ferromagnets, and ferroelastic near MPB region. In particular, the pinning-depinning dynamics caused by randomly distributed defects and impurities is of great interest, since it not only provides fundamental in sight into the physics of driven disordered systems, but is also of technological relevance (e.g., in ferromagnetic and ferroelectric based memory devices) [13,14]. In these materials, domain wall movement can result in a complex nonlinear macroscopic response, displaying significant hysteresis and slow aging. The presence of oxygen vacancies and intrinsic lead vacancies introduces space charge and thereby an internal bias field inside the PZT grains which inhibits the domain motion as a consequence diffuse phase transition are occurred [15, 17]. Here, the diffuse indicates the existence of relaxation process in the material, and the broad dielectric relaxation cannot be described by a single relaxation. This behavior can be associated with compositional fluctuation, which leads to the formation of polar micro or nano-regions with different Curie temperature [18, 19, 20]. Recently, it was reported that oxygen deficient system can stabilize in small fraction of hexagonal unit in cubic system [21, 22]. Such stabilization of two phase system might create nanopolar regions which is responsible for relaxor type of phase transition. It is also exciting to see that these relaxor-type PZT crystals are being used to replace traditional PZT piezoceramics in many new fields outside of medical imaging, future piezoelectric transformers, FeRAM and MEMS actuators [15, 23].

In the present study, we have tried to explain the influence/role of oxygen vacancies in the structural as well as dielectric (relaxor type of nature) properties of Sr^{2+} doped PZT. Polararization studies and ac conductivity also support the presence/influence of oxygen vacancies. In general, $Pb(Zr_{0.5}Ti_{0.5})O_3$(PZT) stabilizes in tetragonal phase, but in present compound presence of Sr^{2+} ions promote the stabilization of oxygen deficient system which support rhombohedral phase formation during the heat treatment. In the present sample, strong electropositive Sr^{2+} ion form a strong bond (bonding energy=426 kJ/mol) with oxygen vacancies rather than Pb^{2+} (bond energy=382 kJ/mol) [7], which plays prominent role in microstructure, electrical and chemical properties of the material. In terms of novelty, author's main focus was to enlighten the consequence of oxygen vacancies on structural and electrical performance of the compound.

Experimental

Polycrystalline sample of $Pb_{0.95}$ $Sr_{0.05}$ $(Zr_{0.5}Ti_{0.5})O_3$ was prepared by a high-temperature solid-state reaction technique using ingredients: $SrCO_3$, ZrO_2, PbO, and TiO_2 (Where used ingredients are $SrCO_3$ is SIGMA ALDRISH purity ($\geq$ 98 %), PbO is LOBA high purity ($\geq$ 99%), ZrO_2 and TiO_2 is LOBA purity ($\geq$ 99.5 %)).

All precursor materials are taken in stoichiometric amount with 3% excess PbO to compensate the lead loss at elevated temperatures and mixed thoroughly in an agate-mortar in a wet condition (methanol) for 4 h. The mixtures were then calcined in a closed alumina crucible. The process of grinding and calcinations was repeated several times to optimize the calcinations temperature. The optimized calcination temperature was 1150 °C. The formation of perovskite phase was confirmed by X-ray diffraction (XRD) technique using an X-ray powder diffractometer (PHILIPS, PW3373 XPERT-PRO). Finally, the calcined powders were used to make cylindrical pellets of diameter 12 mm and thickness 1 – 2 mm under a hydrostatic pressure of about 12 x10^6 N/m^2 using a hydraulic press. Polyvinyl alcohol (PVA) was used as binder which was burnt out during the high temperature sintering process. The pellets were sintered in a covered alumina crucible in air atmosphere for 6 h at 1200 °C. Electrical measurements was carried out in a wide temperature range (25 $\leq$ $T\leq$ 500 °C) using a computer controlled phase sensitive multimeter (PSM_N4L). The hysteresis loops of the poled samples were obtained using workstation of loop tracer, (M/S Radiant Technology Inc, USA).

Results and discussion

Structural and morphological studies

Fig. 1(a) shows the room temperature X-ray diffraction (XRD) pattern of the sample. All the XRD peaks of the system were indexed using tetragonal crystal structure. The diffraction peaks indicate good homogeneity and complete solid solution of $Pb_{0.95}$ $Sr_{0.05}$ $(Zr_{0.5}Ti_{0.5})O_3$ system. The diffraction peaks of Sr doped PZT were fitted with the Vgoit function using a commercial Peakfit program which shown inset **Fig. 1 (a)**.

For the inset **Fig. 1(a)**, these peaks were indexed as (002), (200) in tetragonal phase and (024) in rhombohedral phase. The fraction of rhombohedral phase was estimated using the relation $V_R = I_{R024} /(I_{T200} + I_{R024} + I_{T002})$, where I_{T200} and I_{T002} are the integrated peak intensities of the tetragonal phase and I_{R024} is the integrated peak intensity of the rhombohedral phase. The direct evidence of coexistence of two phases was finally confirmed by Rietveld refinement using X-ray diffraction. The formation of rhombohedral phase could leads to the oxygen vacancy in the system. **Fig. 1(b)** shows the typical scanning electron microscope image of the sintered pellet which is highly dense with grain size

of 5-10 µm range. Beside this the elemental dispersive energy spectrum also shown in **Fig. 1(c)**, which confirms nominal compositions of the present ceramic is homogeneous. The observed X-ray diffraction was refined with tetragonal P4mm space group, which well reported. It was not well fitted with pure tetragonal. Therefore we tried with two phase by using tetragonal phase (space groups; P4mm) along with rhombohedral (space groups; R3c) secondary phases **[24]**. **Fig. 2** represents the Rietveld refinement and difference plot of the calcined powder sample. The most important features (fitting parameters, lattice parameters and R factors) obtained by the results of refinement studies were reported in **Table 1**. Rietveld refinements also conclude that the present system contain major tetragonal phase along with small fraction of rhombohedral one.
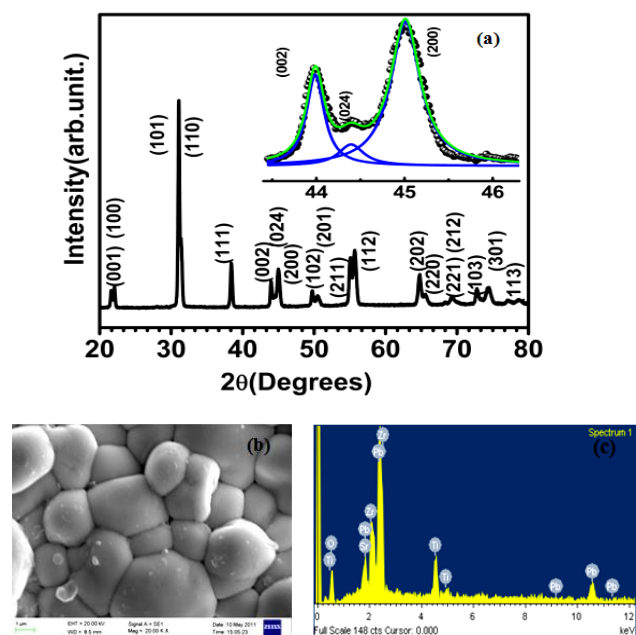

Fig. 1. (a) Room Temperature XRD pattern of the sample, Deconvolution of (002) & 200 peak is shown in inset and (b) SEM micrograph of the sample, (c) elemental dispersive energy spectrum of the sample.
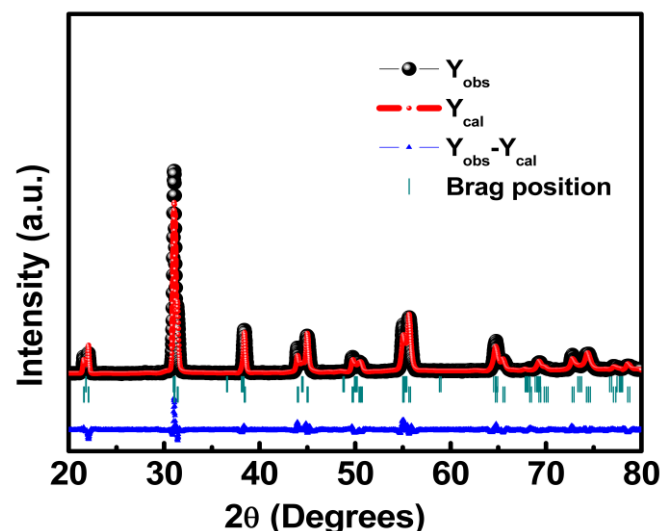

Fig .2. Rietveld refined X-ray diffraction of Sr^{2+} doped PZT.

Table 1. Rietveld quantitative phase analysis.

	Rhombohedral			Tetragonal		
	a = 5.7529 Å, b = 5.7529 Å, c = 14.1650 Å			a = 4.0279 Å, c = 4.1169 Å		
	Rp:16.0 Rwp:15.8 Rexp: 6.44 χ²: 6.05					
	X	y	z	x	y	z
Pb	0.0000	0.0000	0.28300		0.50000	0.52240
Sr	0.0000	0.0000	0.28300	0.50000	0.50000	0.52240
Zr	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
Ti	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
O$_1$	0.13700	0.34700	0.08883	0.50000	0.0000	0.10136
O$_4$				0.0000	0.0000	0.64001

Polarization studies

Fig. 3 shows the polarization v/s electric field (P–E) hysteresis loop with different applied field intensity. The slim and asymmetric loops are observed in the present system. The clearly observed double hysteresis like loop can be explained by the internal stresses in the lattice in a random manner. Hence the internal field may interrupt long range polar order and result in limited polarization under zero field **[25]**. At low frequencies, oxygen vacancies exhibit significant role in electrical properties. The effect of oxygen vacancies and charge carriers is also noticeable in hysteresis plot given in the **Fig. 3**.
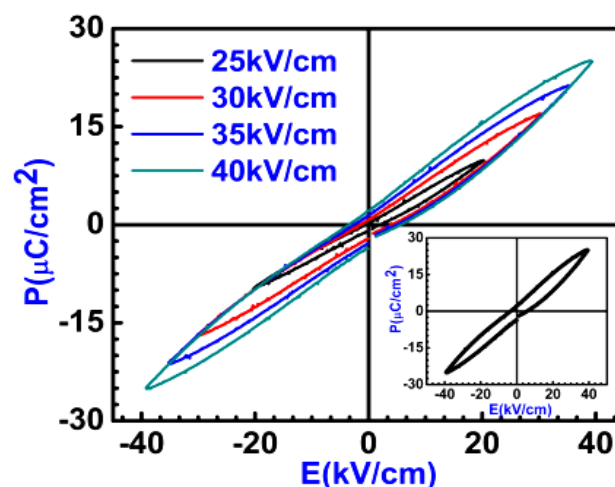

Fig. 3. Room temperature P-E loops of the sample at selective maximum applied field (E$_{max}$), at E$_{max}$ = 40 kV/cm is shown in inset.

At low field, hysteresis loop is not symmetric due to the presence of considerable fraction of oxygen vacancies and charge carrier. The shaping factors of the double hysteresis could be explained in that the ordered defect dipoles lead to the appearance of constricted and double hysteresis loops. This observation can be attributed to the presence of the complex defects, which provide the internal field-bias. Defect-induced lattice deformations can modify local fields and polarization patterns in ferroelectrics **[26, 27]**. Generally, oxygen vacancy is trapped in ferroelectric domain boundaries because these energetically favorable positions act as potential well **[28, 29]**. Defect dipoles formed by the oxygen-vacancy –lattice site associates might be randomly orientated in the system. Such vacancy-

induced antiphase polarizations can play an important role in domain pinning. Ferroelectric ceramics can exhibit the following type of polarization at room temperature [30]:

  i. Polarization associated with highly polarized system contributed by polaron.
  ii. Polarization associated with movement of space charge, cation and anion vacancy.
  iii. Polarization associated with orientation of the switchable ferroelectric domains.

The two former components, corresponding to the nonremanent component, are the most important contributions to the total polarization of the hysteresis loop; whereas, the switchable ferroelectric domains or remanent component contributes in a lesser degree to the loop. Significant differences are observed in the values of P_{max} and P_r for the sample indicates the non-remnant polarization component is dominant. The nonremnent polarization dominancy in present system is due to oxygen vacancy is most probable. The presence of oxygen vacancies stabilized in crystal with different domain orientation and create short range ordering results in nano polar regions induces the relaxor behavior in present sample [25].
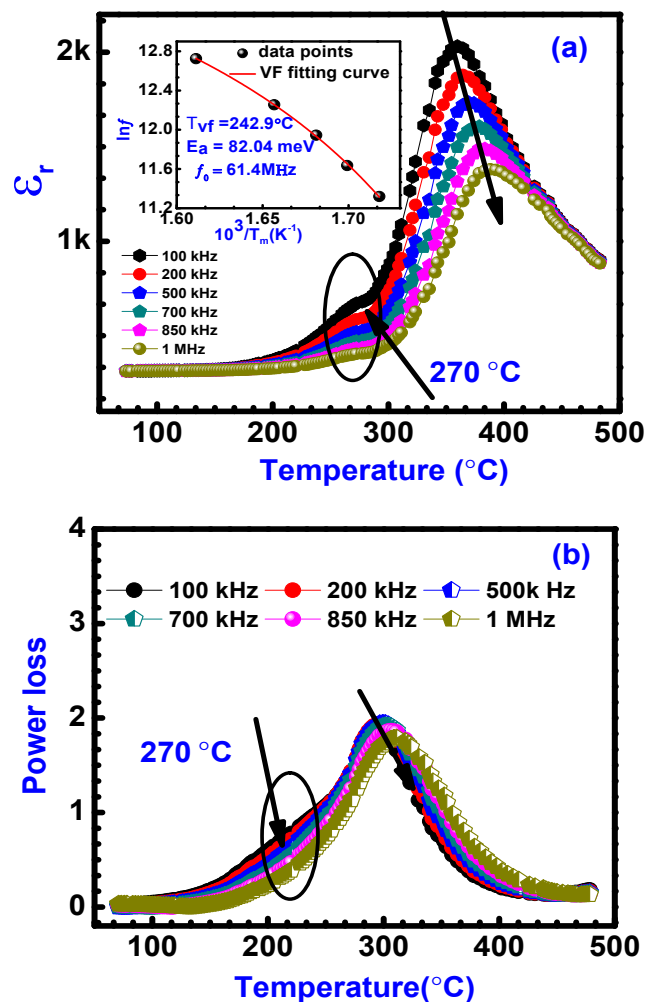

Fig. 4. Variation of relative dielectric permittivity as a function of temperature at different frequencies (100 k Hz -1M Hz).

Dielectric studies

The temperature dependence of relative dielectric permittivity (ε_r) with fixed frequency is shown in **Fig. 4(a)**. It is quite interesting to observe the relaxor behavior in the sample. The large dielectric dispersions are noted below and above T_m which as shift in T_m with increasing frequency from 100 kHz–1 MHz. For understanding the nature of phase transition, we fitted T_m vs frequencies by simple Vogul-Fulcher relation is shown inset **Fig. 4(a)**.

It was confirmed by Vogel-Fulcher relation described as $f = f_o \exp(-Ea/k_B(T_m-T_{VF}))$. Which can explain a freezing process of polar nanoregions (PNRs), where f is the measured frequency, f_0 is a pre exponential function, E_a is activation energy, k_B is the Boltzmann constant, T_m is the temperature corresponding to the dielectric maxima, and T_{VF} is the characteristic Vogel–Fulcher freezing temperature. The fitted parameters are shown inset **Fig. 4(a)** are well agreed with previous reports [31]. At certain nonzero temperature thermal motion is capable of destroying the ordered oxygen sites may create polar nanoregions inside the grains. Due to such orderly arranged PNRs can acts as unusually large dipoles are always frequency dependent on an external electric field. Variation of dielectric parameters (ε_r and tanδ) with temperature also confirms the existence of two phases in the prepared compound. In **Fig. 4(a)**, first phase transition is observed over the temperature range 260-270 °C which is evidence of rhombohedral to tetragonal phase transition and second one is evidence of ferroelectric tetragonal to paraelectric cubic phase over the temperature range 350-390 °C. Such kind of behaviour has also been reported earlier [31-33]. Moreover, the dielectric loss also shows similar trend as dielectric permittivity. It was quite high like normal relaxor materials [34, 35] are shown in **Fig. 4(b)**. Similar frequency dependent loss also noted at same temperature range.

Conductivity studies

The ac conductivity (σ_{ac}) of the system was calculated using the relation $\sigma_{ac} = \omega\varepsilon_0\varepsilon_r\tan\delta$ (ω = angular frequency, and ε_0 = dielectric constant in free space) from the measured dielectric parameters. The variation of ac conductivity (σ_{ac}) with $10^3/T$ at 10k Hz is shown in **Fig 5**. The conductivity study on the dielectric specimen at different temperature range can provide total real charge carrier (i.e. conducting species) in the sample. From the graph the major conclusion drawn, as it has three regions; three conduction mechanisms associated with three regions in materials.

(1) At low temperature electron hoping occurs between two lattice site. In the present system, we have denoted as region 3 with activation energy of 0.158eV. In fact in perovskite ferroelectric materials oxygen vacancies are considered as one of the mobile charge carriers through formation of singly and/ doubly ionized oxygen vacancies with the release of single or two electrons respectively from the oxygen lattice site. During these creation processes, some of the released electrons are captured by Ti^{4+} to form Ti^{3+}, The electronic hopping between ($Ti^{3+} – Ti^{4+}$) state leads to over all electronic conductivity at low temperature [36].

(2) In region 2, near phase transition the observed activation energy is 0.605 eV. The lattice strain originated during the increment of the temperature below the transition temperature, then crystal attains polar mode. The electrons interact with the polar modes of the crystals to form small polaron, which is responsible for conduction process in transition region [37]. Hopping process transports the carriers.

(3) In region 1 which was above phase transition, the motion of oxygen vacancies is not localized in one unit cell but can be extended to the whole system leading to ionic conductivity. The ionic conductivity is an intrinsic and typical conduction mechanism in ferroelectric materials at high temperatures [38]. Therefore, each of the above mentioned mechanisms and the electrical conductivity are thermally activated.
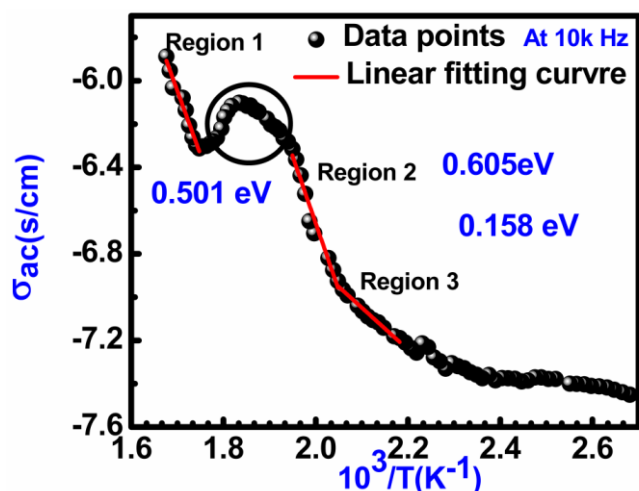
Fig. 5. Variation of ac- conductivity (σ_{ac}) with inverse of absolute temperature (1000/T) at 10 kHz.

Conclusion

In summary, we have been carried out structural, microstructural and elemental compositional analysis of $Pb_{0.95}Sr_{0.05}(Zr_{0.5}Ti_{0.5})O_3$ ceramic. It is also shown that diffuses relaxor type behavior in dielectric studies and further confirmed by Vogul-Fulcher relation. Dielectric property (relaxor behavior) is significantly influenced by oxygen vacancies and corresponding evidence of the existence of oxygen vacancies was also proved by the conductivity and polarization results.

For future prospective, present study is very useful to identify the existence of oxygen vacancy and proper study of its influence on structural, electrical and polarization properties.

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