

# Structural, dielectric and piezoelectric properties of aluminium doped PLZT ceramics prepared by sol–gel route

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## Abstract

Nanosized piezoelectric ceramics for vibration sensor applications have been prepared by mixing the ferroelectric PLZT (8:60:40) with variable doping fractions of trivalent aluminium ion ( $\text{Al}^{3+}$ ). Samples have been prepared through a standard sol–gel route. X-ray diffraction and scanning electron microscopy (SEM) have been used to determine the phase and morphological modifications. Transmission electron microscopy (TEM) studies reveal the microstructure with nanosized well-dispersed homogeneous spherical particles. The vibrational infra-red (IR) spectroscopy record is taken to locate the position of the doping  $\text{Al}^{3+}$  ion. Using electrical impedance spectroscopy, the resonance and anti-resonance frequencies of the Al modified PLZT system have been determined and analysed. Al addition in PLZT has left a profound effect in its dielectric and piezoelectric properties. An interpretation of the role of Al addition is proposed in terms of structure modification. The sensing power of the investigated material was found useful for the vibration control of a cantilever beam.

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## 1. Introduction

Since the discovery of ferroelectricity in single-crystal materials (Rochelle salt) in 1921 and its subsequent extension into the realm of polycrystalline ceramics ( $\text{BaTiO}_3$ ) during the early to mid 1940s, there has been a continuous succession of new materials and technology for commercial applications. The growing interest in search of suitable materials has led to the development of a large number of new complex ferroelectric ceramics with a wide variety of composition and stable structure. Materials based on two systems, barium titanate ( $\text{BaTiO}_3$ ) and lead zirconate titanate (PZT), have dominated the field of ferroelectrics throughout its history. One system that embraces all compositional aspects of the dielectric, piezoelectric, pyroelectric, ferroelectrics and electrooptic ceramic is the La modified PZT (PLZT). PLZT is a perovskite ferroelectric formed by doping  $\text{La}^{3+}$  ions at the A-site of lead zirconate titanate (PZT). It is well known that a suitable ionic substitution

at different atomic sites of  $\text{ABO}_3$  type materials has a dramatic effect on their physical properties. It has been shown that the chemical substitutions [1–4], the addition of oxides [5] and the sample shape [6] might have a prominent influence on the structure of PZT, and on electrical and mechanical properties of the PZT based ceramics. Depending upon the specific requirements for different applications of piezoelectric ceramics, various compositions of the Zr/Ti ratios may be chosen. For instance, a material with high value of  $k_p$  and permittivity a composition near MPB is chosen [7] whereas for a material with high mechanical quality factor  $Q_m$  and a low permittivity a composition far away from the MBP is preferred. Keeping in mind the practical applications of PLZT various impurities have been added with a favored composition of La:Zr:Ti in close proximity of MBP (8:65:35 and 8:60:40) in order to improve the desired device parameters. In view of this, we have synthesized and studied various physical properties of a group of new compounds with trivalent doping (Fe, Sb, Al, Bi, Ga) in PLZT [8–12], prepared through different routes. It is known that the crystallite size plays an important role in determining the physical properties. So for better homogeneity we have prepared the samples for the present investigation through

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a sol–gel technique, which provides molecular level mixing of the elements, which decreases the diffusion path (up to a nanometric scale) for getting the required material in nanoscale [13]. In our present investigation the structural and electrical properties of a group of piezo ceramics with a general formula  $[\text{Pb}_{0.92}(\text{La}_{1-z}\text{Al}_z)_{0.08}(\text{Zr}_{0.60}\text{Ti}_{0.40})_{0.98}\text{O}_3]$ ;  $z = 0.0, 0.3, 0.6, 0.9$  and  $1.0$  are studied which were prepared through a sol–gel route in nanorange by satisfying the following conditions: (a) charge neutrality and (b) Goldschmidt's tolerance factor,  $t$  [10,14,15]. In our earlier study [10], we reported the effect of same dopant (Al) on structural and dielectric properties of PLZT, prepared via high temperature mixed oxide reaction technique. Since in the present investigation materials are prepared in nanoscale, we have repeated the studies associated with structural and dielectric properties to observe the nanosize effect on those properties. We have also carried out some additional structural property studies; e.g., TEM to confirm the particle size and IR study to investigate the positional change of atomic spectra with doping ion. In this paper we have mainly focused on piezoelectric properties and tried to make use of the samples as a device in smart structures.

## 2. Experimental

Polycrystalline samples of  $[\text{Pb}_{0.92}(\text{La}_{1-z}\text{Al}_z)_{0.08}][\text{Zr}_{0.60}\text{Ti}_{0.40}]_{0.98}\text{O}_3$  ( $z = 0.0, 0.3, 0.6, 0.9, 1$ ) (hereafter PLAZT) were prepared by a sol–gel technique following the procedure/steps reported elsewhere [16,17]. Finally, the calcined powder was used to make cylindrical pellets under an uniaxial pressure of about  $6.5 \times 10^7 \text{ N/m}^2$  using a hydraulic press. The pellets were sintered in a covered alumina crucible at  $1250^\circ\text{C}$  in a  $\text{PbZrO}_3$  atmosphere for 24 h. In order to prevent Pb/PbO loss and to maintain the stoichiometry of the compounds during sintering at high temperature, an equilibrium  $\text{PbO}$  vapour pressure was established with  $\text{PbZrO}_3$  as setter by placing everything in a covered platinum crucible. The formation of the desired compounds was checked by preliminary structural analysis using X-ray diffraction (XRD) pattern recorded on calcined powder in a wide range of Bragg angles,  $2\theta$  ( $20^\circ \leq 2\theta \leq 80^\circ$ ) at room temperature with an X-ray diffractometer Philips PW1710, Holland by  $\text{Co K}\alpha$  radiation ( $\lambda = 1.7902 \text{ \AA}$ ). The peak broadening due to mechanical strain, instrumental and other means has been ignored since powder samples were used. The surface morphology/grain distribution of the pellet surface was studied using JEOL JSM-5800 scanning electron microscope (SEM) at room temperature with different magnifications. The microstructures of the pellet samples were studied by transmission electron microscopy (HITACHI H-600, Electron Microscope) to calculate the actual particle size of the compound.

Fourier transform infra-red (FTIR) spectra of PLAZT ( $z = 0.0$ – $1.0$ ) were recorded using a computer controlled THERMO NICOLET FTIR spectrophotometer (Model: Nexus-870) in the wave number range  $4000$ – $400 \text{ cm}^{-1}$  at room temperature ( $-25^\circ\text{C}$ ). The experiment was carried out in an inert argon atmosphere by averaging 32 scans with a speed of 2 s per scan. The optical resolution for all spectra was  $4 \text{ cm}^{-1}$ .

The dielectric measurements were obtained on electroded-sintered pellet and the data were recorded using computer-controlled LCR (HIOKI 3532) impedance bridge, over a wide range of temperature ( $30$ – $500^\circ\text{C}$ ) at a dc signal of 1.3 V.

Electromechanical properties were determined following piezoelectric strain coefficient  $d_{33}$  of poled ( $12 \text{ kV/cm}$  dc at  $100^\circ\text{C}$ ) samples using a Piezo-Meter (PM35 Take-Control, UK) at 100 Hz (pressure 1 N) and room temperature. Electromechanical coupling coefficient ( $k_p$ ) of the samples were calculated from the resonance ( $f_r$ ) and anti-resonance ( $f_a$ ) frequency from a set of measured impedance data using the formula:

$$\frac{k_p^2}{1 - k_p^2} = 2.51 \frac{f_a - f_r}{f_r}$$

To investigate the sensing and actuation property of the prepared samples and use it as a vibration sensor a simple apparatus has been fabricated with a vibratory load, and a computer-controlled piezo-sensing instrument. For this measurement poled piezo patches were embedded into the root of the carbon composite cantilever beam. The cantilever specimen was rigidly fixed at the root with the clamps. The tip of the shaker was kept touching the cantilever very near the root end. Function generator output was fed to power amplifier input and the power amplifier output was fed to the shaker. The connecting wires from the piezo acting as the sensor were connected to the piezo-sensing system inputs. The output of the piezo-sensing system was then fed to the band pass filter system for filtering out the noise present. The output of the band pass filter system was fed through connecting wires to the piezo actuation system. The output of the piezo actuation system was then fed to the piezo acting as actuator via connecting wires. The damped out response and closed loop vibration patterns of the beam with and without the samples were observed in an oscilloscope and were recorded by the computer.

## 3. Results and discussion

The room temperature XRD pattern of the PLAZT compounds for all the Al concentration is shown in Fig. 1. All the observed peaks/reflections are indexed in tetragonal crystal systems on the basis of the best agreement between observed and calculated  $d$  values. The unit cell constants obtained for the tetragonal phase at room temperature are in good agreement with previous work on doped PLZT (8:60:40) [10,18]. A comparative study on tetragonality and unit cell volume with doping level of Al concentration is given in Fig. 2. Fig. 3 shows SEM micrograph of pure and modified PLZT. If we compare the micrograph of doped PLZT (i.e.,  $z = 0.3$ – $1.0$ ) (Fig. 3b–e) with pure one  $z = 0.0$  (Fig. 3a) we can see the presence of alumina immersed into the major PLZT phase. A certain degree of porosity is observed by incorporation of higher percentage of  $\text{Al}^{3+}$  ions. TEM (with SAED pattern in inset) micrographs are shown in Fig. 4a and b for  $z = 0.0$  and  $1.0$  samples. Particle size calculated from TEM is found to be  $\sim 12$ – $20 \text{ nm}$ .

In order to confirm the substitution of smaller cations like Al, at the Pb/La site (A-site of  $\text{ABO}_3$ ), the vibrational spectroscopic experiment was carried out in the wave number region of interest  $400$ – $100 \text{ cm}^{-1}$  (Fig. 5). The spectral pattern of the typical

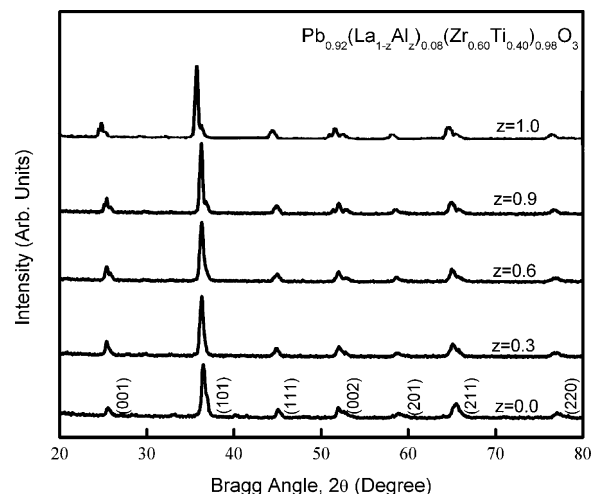


Fig. 1. Room temperature XRD pattern of PLAZT ceramics.

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