

The relationship between ferroelectric domain pattern and properties of PLT21 ceramics

F A Londoño¹, J A Eiras² and D Garcia²

¹ Universidad de Antioquia, Medellín, Colombia.

² Universidade Federal de São Carlos, São Carlos, Brasil.

E-mail: fernandoa.londono@udea.edu.co

Abstract. The light passing through a ferroelectric crystal tends to split into several separate beams due to refraction and reflection at the domain walls. This interaction of light with ferroelectric domains becomes one of the most important features of modern optoelectronic and nonlinear optics materials. Recently, lanthanum modified lead titanate, $\text{Pb}_{(1-x)}\text{La}_x\text{TiO}_3$, (PLT) has become popular because it possesses interesting properties such as a lower Curie temperature, a lower coercive field, and smaller remanent polarizations than PZT and has great potential for nonlinear optical and electro optical applications. In this work, we propose a simple model taking into account the optical transmission and rearrangement of the ferroelectrics domains structure on the PLT21 ceramic in temperature function. The variation of dielectrical and optical measurements in function of temperature were observed and correlated with the optical visualization of ferroelectric domains.

1. Introduction

Lead titanate PbTiO_3 (PT), a ferroelectric material, is known for its interesting properties; high Curie temperature, pyroelectric coefficient, and spontaneous polarization, and low dielectric constant. These properties make it suitable for numerous applications: ultrason transducers, thermistors, optical electronic devices etc [1]. At room temperature, PT has a tetragonal perovskite structure, and combined with other oxides it forms materials such as $(\text{Pb},\text{La})\text{TiO}_3$, (PLT) [1,2]. $\text{Pb}_{(1-x)}\text{La}_x\text{TiO}_3$ is a ferroelectric system with excellent dielectric and piezoelectric properties [3]. However, this material can also be obtained as transparent ferroelectric ceramics for relatively high La content, $0.18 \leq x \leq 0.22$ [4, 5]. Transparent ferroelectric ceramics (TFC) have recently acquired a high interest due to their potential application as electro optics devices [6]. The degree of transparency of TFC decrease due to extrinsic defect (as example, pores and secondary phases at the grain boundaries) and intrinsic defect (as example, ferroelectric domains pattern) [7]. Therefore, the main objective of this work was to correlate the optical and electrical response with domain pattern behavior as a function of the temperature for the ceramics $\text{Pb}_{(1-x)}\text{La}_x\text{TiO}_3$ with $x=0.21$ (PLT21), (PLT21 has major transmittance than the other compositions [4]) prepared through the solid-state reaction method.

2. Experiments

The raw materials PbO purity (99.3%), TiO_2 (99.9+%), and La_2O_3 (99.9%), were weighed according to the formula $\text{Pb}_{(1-x)}\text{La}_x\text{TiO}_3$, with $x=0.21$. The primary oxides were ball-milled



in distilled water, with zirconia as the grinding medium, for 3h. The resulting slurries were then dried at 120°C and later calcined at 850°C for 3h in air. The calcined powders were ball-milled in distilled water for 3h and dried again. Binder was added to the powders, which were uniaxially cold-pressed into disks (10mm in diameter and 10mm thick), labeled according to nominal composition, PLT21 ($x=0.21$). Subsequently, isostatic pressure was applied to reduce the density gradients. After burning out the binder, the disks were hot-pressing at 1220°C, for 3h, under a uniaxial pressure of ~ 6.0 MPa, in O_2 atmosphere (30kPa).

The purity of the phases and the structural parameters were determined by X-ray diffraction (XRD) using a Rigaku diffractometer, with CuK radiation. The lattice parameters were calculated by least-square fitting from the positions of the diffraction peaks. The ideal density was calculated from the unit cell volume found by XRD analysis and the molecular weight. For dielectric measurement, gold electrodes were deposited on both faces of the disk samples (5 mm diameter and ~ 1.0 mm thick). The relative permittivity was measured by impedance analysis (HP4194A). The transmittance was measured in a spectrophotometer (Micronal-B582), at wavelengths ranging from 200 to 1000nm for optically polished samples, 600m thick [8]. The visualization of the domains was carried out with a microscope Nikon, Eclipse ME600, interfaced to a control system of temperature Linkam (room temperature to 200°C). The microscopic patterns are studied using transmittance light. Magnifications in the 100X to 200X range were realized for domain visualization in ceramics with thickness 0.1mm width 4mm and length 5mm.

3. Results

The results revealed, homogeneous microstructure, grain size of $(10.3 \pm 0.8)\mu m$, high optical transmittance and high relative density (97.8%) [4, 7]. In the Figure 1(a) can be seen DRX patterns of PLT21 ceramic. It is observed and calculated the absence of second phases with perovskite structure and tetragonality factor, $c/a=1.013$. The anisotropy as measured by the tetragonality of the unit cell, c/a can be considered interest for electrical properties studies [9].

The temperature profile of real relative permittivity under frequency of electric field 1kHz for PLT21 is shown in Figure 1(b). The value of relative permittivity ϵ' is increased with increasing temperature and reach a maximum value at 129° C, then it is decreased with increasing temperature. High dielectric permittivity and only transition can be seen. This measurement is typical of material ferroelectrics normal with phase transition (ferro-para transition) [9, 10].

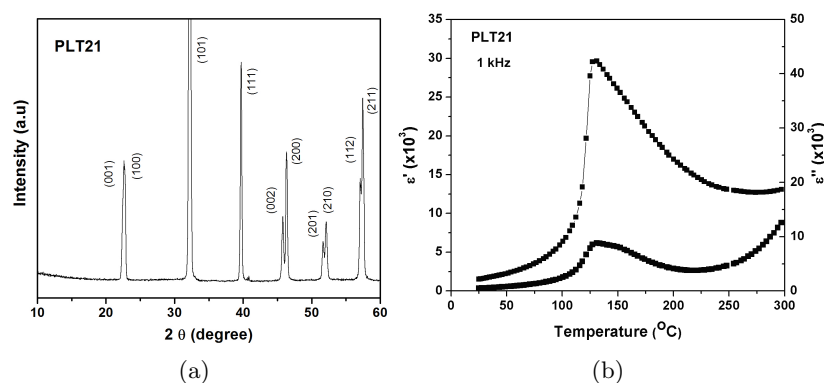


Figure 1. (a) XRD patterns of PLT21 ceramics to room temperature, (b) temperature dependence of ϵ' and ϵ'' , measured at 1kHz for PLT21 ceramic.

Figure 2 shows optical micrograph to different temperatures of PLT21 ceramic. Also in these figure is inserted the dielectrical measurements in function of temperatures. In the first micrograph can be observed clearly many 180° domains under polarized light to 27°C, however, the domains disappear with the increase of temperature. In the same way, for temperatures higher than the the phase transition temperature (129°C) it is can be seen

as the ferroelectric domain disappears completely, however, the domains reappear when the temperature decrease. This observation can be used for the relationship between ferroelectric domain pattern and dielectrical properties of PLT21 ceramics, especially in determining the phase transition temperature of normal ferroelectrics materials.

The most important result of this observation is that we can easily and quickly distinguish ferroelectric domains of PLT21 ceramics.

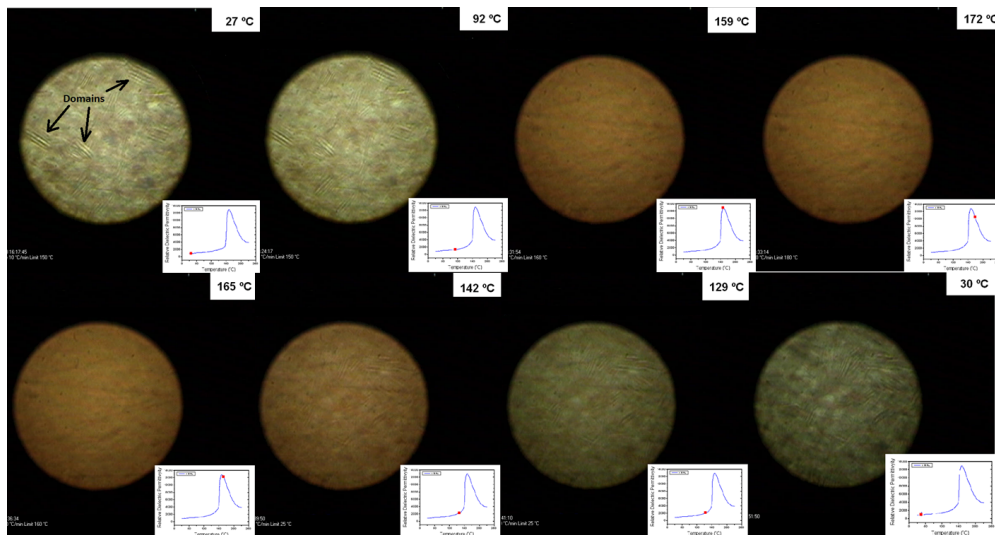


Figure 2. Relationship between ferroelectric domain pattern and dielectrical properties of PLT21 ceramics.

4. Conclusions

The assembly described in this paper operates as a light beam deflector. The deflection is accomplished by total internal reflection of the beam at the boundaries of a 180° domain wall in a ceramic and can be used for the determination of phase transition temperature in normal ferroelectrics materials.

This work is the first in the optical visualization of ferroelectric domains of the system PLT21.

5. Acknowledgements

This research was partially supported by Colombian Agencies: CODI-Universidad de Antioquia (Estrategia de Sostenibilidad 2014-2015 de la Universidad de Antioquia and project: “Síntesis y caracterización de cerámicas transparentes multifuncionales para aplicaciones en dispositivos electro-ópticos y para la construcción de matrices para láseres de estado sólido”), FaCEN-Universidad de Antioquia and to CAPES, FAPESP and CNPq for the financial support.

References

- [1] K Limame and M Kellati and S Sayouri 2005 *Moroccan Journal of Condensed Matter* **6-1** 44
- [2] D Garcia, F A Londono and J Eiras 2010 *Revista Latinoamericana de Metalurgia y Materiales* **31-1** 52
- [3] Vasco E, Vzquez L and Zaldo C 1999 *Applied Physics A* **69-1** S827
- [4] D Garcia, F A Londono and J Eiras 2012 *Boletn de la Sociedad Española de Cerámica y Vidrio* **51-6** 353
- [5] Yamamoto T, Igarashi, Hideji, Okazaki, Kiyoshi 1983 *Journal of the American Ceramic Society* **66-5** 363
- [6] A Sternberg 1989 *Ferroelectrics* **91-1** 53
- [7] D Garcia, F A Londono and J Eiras 2012 *Optical Materials* **34** 1310
- [8] J Eiras, F Milton, F Londono and D Garcia 2012 *Optics and Photonics Journal* **2-3** 157
- [9] G Shirane and S Hoshino 1952 *Journal of the Physical Society of Japan* **6-4** 265
- [10] Vijendra A Chaudhari and Govind K Bichile 2013 *Smart Materials Research* **2013-147524** 1 (doi:10.1155/2013/147524)