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Microstructure and Dielectric Properties of PZS-PLZT Ceramics System

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ABSTRACT

Piezoelectric ceramics are an important class of solid-state materials as they exhibit a wide range of applications. This article investigates the sintering temperature effect on microstructure and dielectric properties of a new ceramics material Pb (Zn, Sb) O_3-PbLa (Zr, Ti) O_3 (PZS-PLZT), which was prepared by a solid mixed-oxide solution method. Scanning electron micrograph (SEM) and X-ray diffraction (XRD) techniques was employed to examine the crystallization of the ceramics. The results of XRD show that the phase structure of the samples is tetragonal. The lattice parameters and the density increased with increase of the sintering temperature. The dielectric constant and the dielectric loss of the investigated samples decreased with increase in the frequency.

Keywords: Ferroelectric; Microstructure; Piezoelectric Ceramics; Dielectric Properties

1. Introduction

The piezoelectric property plays an important role for electric and electronic materials. The most widely used piezoelectric materials are based on the $PbTiO_3$-$PbZrO_3$ system (PZT) [1-3]. The application of the PZT component is wide, including nonvolatile memory elements, pyroelectric detectors, piezoelectric transducers, and photoelectric devices [4,5], but the electrical and thermal properties of PZT ceramics depend to a large extent on their surface microstructures [6]. Many researchers have considered modified PZT compositions with suitable substitution at the A and B sites [7-9] with some dopants to improve their electrical and mechanical property of piezoelectric materials in recent years, this dopants can be classified into two categories: donor dopants which induce Pb vacancies by changing with a higher valence ion for Pb^{2+} or $(Ti, Zr)^{4+}$ like Sb^{5+}, La^{3+}. And the acceptor dopants induce O vacancies by changing with a lower valance ion for Pb^{2+} or $(Ti, Zr)^{4+}$ example K^+, Fe^{3+} [10, 11]. So the properties of a PZT samples are very sensitive to additives, composition and synthesis methods.

Whatmore *et al.* [12] and Chaipanich *et al.* [13] have reported that the electric properties of PZT ceramics could be improved by the doping of Sb_2O_3. [14]. It was found that sintering behavior had obvious effects on the microstructure and piezoelectric properties. With the increase of sintering temperature, the microstructure of the ceramic samples changed accordingly and the porosity decreased [15].

There have been several reports on La substituted PZT. It has been reported that substitution of Pb^{2+} by La^{3+} ions created vacancies in the A site of perovskite ABO_3 structured PZT ceramics. The 7% La doped PZT ceramics PLZT (7/60/40) composition is one of the most important candidates for piezoelectric applications due to its extremely high piezoelectric and electromechanical coupling coefficients reported in literature [16-18].

In this work, a new ceramics material based PZT is prepared by a solid mixed-oxide solution method, and we investigate the structural, dielectrical properties of a doped PZT ceramic with La, Sb and Zn for different sintered temperature.

2. Experimental

The composition of PZS-PLZT system was $0.3Pb(Zn_{1/3}, Sb_{2/3})O_3$-$0.7Pb_{0.98}La_{0.02}(Zr_{0.48},Ti_{0.52})O_3$ were synthesized from high purity oxide powders PbO (99.90%), ZrO_2 (99.90%), TiO_2 (99.80%), Sb_2O_3 (99.90%), ZnO (99.90%), and La_2O_3 (99.90%), the samples were prepared by the conventional ceramic procedure. Stoichiometric amount of metal for the designated PZS-PLZT

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composition was mixed for 2 hours as the grinding media and ethanol as the solvent. After milling for 6 hours, the resultant slurry was dried in an oven and then the powders were calcined in a high-temperature furnace at 850˚C for 120 min. Then, the calcined powder was ball-milled again to ensure a fine particle size for 6 hours. After drying, the powder was pressed as disks and sintered at two different temperatures 1050˚C and 1200˚C in a closed aluminum crucible, the atmosphere was enriched in PbO vapor using $PbZrO_3$ powder, the density of the sintered component was measured from its mass and dimensions. The sintered discs were next mechanically processed by abrasion, dried and electrified with a silver paste burned out at 700˚C for 30 min. The polling was carried out in a silicon oil bath at 100˚C under an electric field of 3 kV/mm and cooled down to room temperature still being under the influence of the electric field. They were aged for 24 h prior to testing. The temperature dependence of dielectric properties was measured at temperatures ranging from room temperature to 650˚C with a heating using an impedance analyzer. An X-ray diffract meter BRUKER-AXE, D8 with CuKα radiation (λ = 1.5406 Å). Using the JCPDS database we can identify the presence of the perovskite structure and phases of the sintered samples. Surface microstructures of the ceramics were observed using a scanning electron microscopy (SEM) (JEOL JSM-6390LV) at room temperature.

3. Results and Discussion

3.1. X-ray Diffraction (XRD)

The XRD patterns of PZS-PLZT ceramics shown in **Figure 1** were identified as a material with perovskite structure having tetragonal symmetry according to JCPDS (Joint Committee of Powder Diffraction Standards) card n 00-046-0504. The tetragonal lattice parameters were determined from the evolution of the tetragonal peaks (200) and (002) by using *CELREF* software.

The results given in **Table 1** revealed that the tetragonal cell parameters are associated with some physical properties of the sintered samples. The lattice parameters and the density increased with increase the sintering temperature.

3.2. Scanning Electron Micrograph (SEM)

Figure 2 shows the scanning electron micrograph (SEM) microstructure of the porous PZS-PLZT prepared samples sintered at 1050˚C and 1200˚C by (SEM: JEOL JSM-6390LV). The increase of grain size from ≈1.83 μm to ≈3.12 μm is observed significantly with the increasing of sintering temperature from 1050˚C to 1200˚C respectively, and it can clearly be seen that the prepared PZS-PLZT ceramics is porous and has a not uniform distributions of grain shape and size.

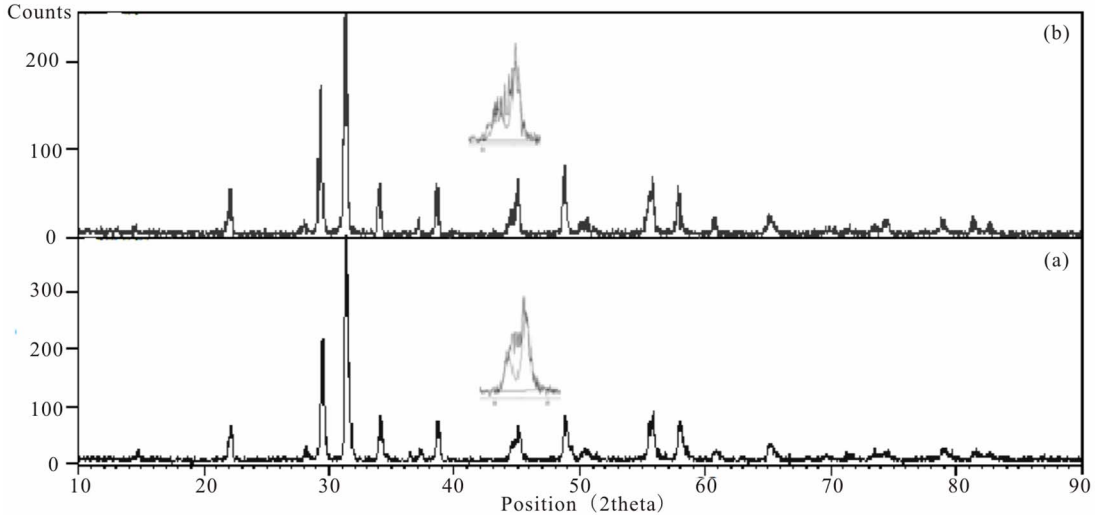

Figure 1. XRD patterns of PZS-PLZT ceramics sintered at (a) 1050˚C and (b) 1200˚C.

Table 1. Some physical properties of the sintered PZS-PLZT ceramics.

Sintering temperature (˚C)	Thickness [cm]	Diameter [cm]	specific weight [g/cm^3]	phase	lattice parameters		
					a [Å]	*b* [Å]	*c* [Å]
1050˚C	0.18	1.20	6.635	T	4.0155	4.0155	4.0517
1200˚C	0.18	1.15	7.598	T	4.0346	4.0346	4.0714

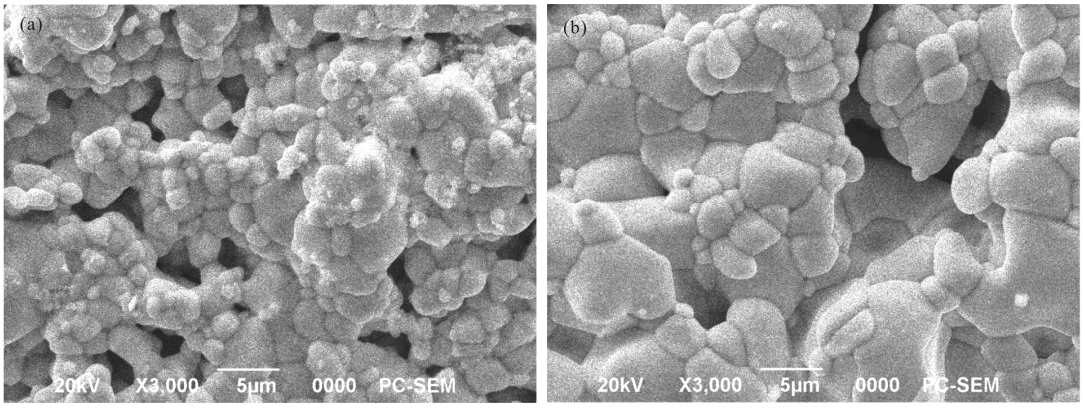

Figure 2. SEM micrographs of PZS-PLZT ceramics sintered at (a) 1050˚C; (b) 1200˚C for 2 hours.

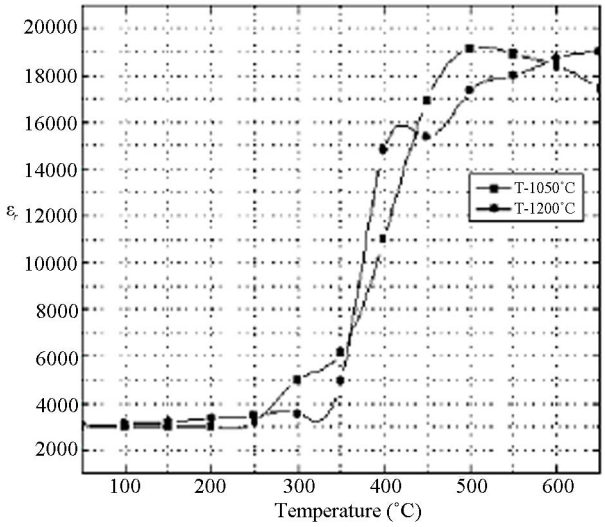

Figure 3. Temperature dependence of ε_r for PZS-PLZT samples sintered at 1050˚C and 1200˚C.

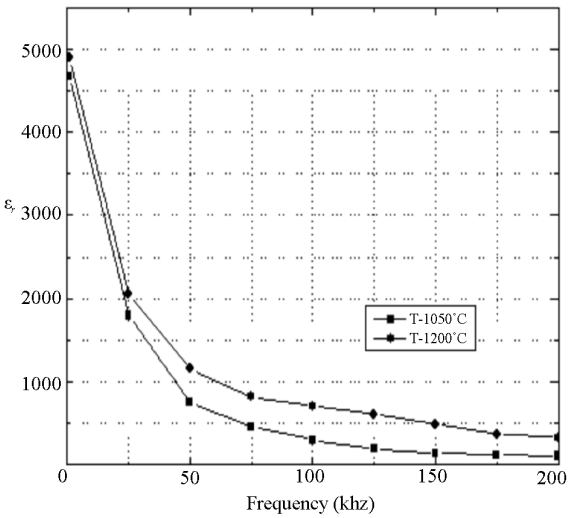

Figure 4. Frequency dependence of ε_r for PZS-PLZT samples sintered at 1050˚C and 1200˚C.

3.3. Dielectric Properties and the Loss Factor

Figure 3 shows the variation of the dielectric constant at 5 kHz with increment of temperature for the two samples witch sintered at 1050˚C and 1200˚C. Similar to normal ferroelectrics, the dielectric constant increases gradually with increasing temperature up to the transition temperature, then it decreases. Here we can note the most important characteristic of a disordered perovskite structure. The dielectric constant reaches a maximum value of 19,130 (T_c = 500˚C) for sample sintered at 1050˚C, but for the sintering temperature 1200˚C, the dielectric constant continues increasing (out of range temperature T_c > 650˚C). There, may be that this result is due to the change in the bulk density and the grain morphology with the change of sintering temperature [19]. This constant decreases gradually with increasing frequency (**Figure 4**) it reaches a maximum value at small values of frequency which explained by existence of different polarization models [20,21]. The dielectric constant of

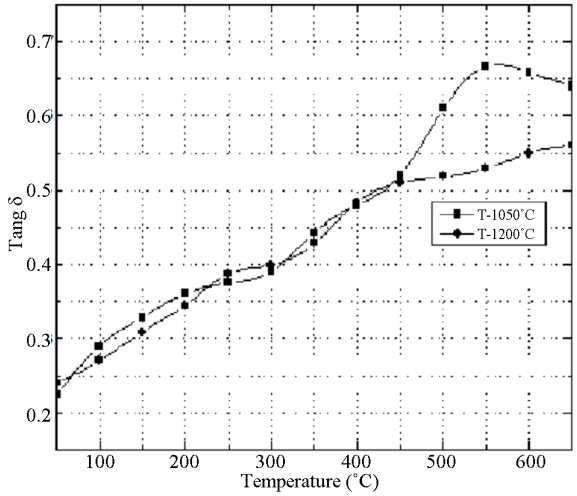

Figure 5. Temperature dependence of Tangδ for PZS-PLZT samples sintered at 1050˚C and 1200˚C.

sample sintered at 1200˚C decreases slowly than of

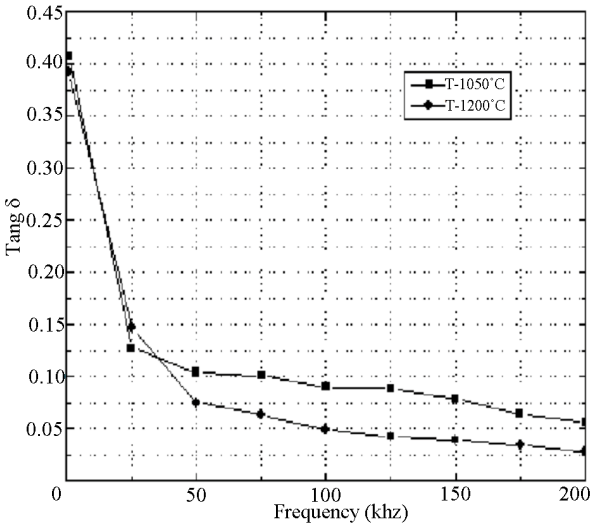

Figure 6. Frequency dependence of Tangδ for PZS-PLZT samples sintered at 1050˚C and 1200˚C.

1050˚C sintered sample.

Figure 5 shows the variation of the loss factor *Tang*δ with temperature. Tangδ increases gradually with increasing temperature. For sintered temperature 1050˚C the loss factor reaches a maximum value of 0.65 at (T_c = 550˚C), but for the sintering temperature 1200˚C, the dielectric constant continues increasing (out of range temperature T_c > 650˚C). This may be caused by the losses due to the electrical conduction. This factor decreases with increasing frequency (**Figure 6**), which is a characteristic of ferroelectrics [22,23].

4. Conclusion

PZS-PLZT was successfully prepared by a solid mixed-oxide solution method; The X-ray diffraction measurements for all $0.3Pb(Zn_{1/3},Sb_{2/3})O_3$-$0.7Pb_{0.98}La_{0.02}(Zr_{0.48}, Ti_{0.52})O_3$ sintered samples indicated the presence of tetragonal phase and the scanning electron micrograph shows a porous nature of ceramics and the grain size increases with increasing the temperature. The influence of the sintering temperature on the properties of PZS-PLZT ceramics was studied. The lattice parameters and the density increased with increase of the sintering temperature. The dielectric constant and the dielectric loss of the sintered samples decreased with increase in the frequency. The effects of the sintering temperature on the dielectric properties can be attributed to the change in the bulk density and the grain morphology with the change of the sintering temperature.

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