Effect of Sb_2O_3 Doping $Pb(Zr_{0.53}Ti_{0.47})O_3$ Ceramics

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Abstract

$(Pb_{1-x}Sb_x)(Zr_{0.53}Ti_{0.47})O_3$ ceramics when x = 0.0, 4.5, 6.0 and 7.5 mol % were synthesized and measured dielectric and ferroelectric properties. Which the powder of PbO, ZrO_2, TiO_2 and Sb_2O_3 were mixed by ball – milling method and calcined at 1123 K for 4 h and the sintered at 1423 K for 2 h in air. The crystal structure was analyzed by XRD technique; the crystal size was identified by Scherer's equation and calculated the theoretical density. The crystal size is 267.29 Å, 292.11 Å, 686.05 Å and 642.89 Å, the theoretical density is 7.51 g cm^{-3}, 7.41 g cm^{-3}, 7.22 g cm^{-3} and 7.45 g cm^{-3}, the Vickers hardness is 425.40 Nmm^{-2}, 496.20 Nmm^{-2}, 572.20 Nmm^{-2} and 614.60 Nmm^{-2} when x = 0.0, 4.5, 6.0 and 7.5 mol % respectively. Addition with Sb in the same position of Pb it is another alternative in the development property of PZT (53/47) ceramics and improving properties of dielectric and ferroelectric for PZT (53/47) ceramics.

KEYWORDS: Crystal structure; Piezoelectric Ceramics; Dielectric; Ferroelectric

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Introduction

According to the study scientists at the conducted studies on the piezoelectric materials there are two groups is lead zirconate titanate (PZT) and free – lead. PZT ceramics has special ferroelectric properties and is widely used for power codec applications. PZT ceramics often be corrected with the addition on the A site and B site of Perovskite structure for the soft and hard PZT materials [1 – 3]. The composition of PZT which has Zr/Ti ratio ≈ 53/47 show high ferroelectric and dielectric properties due to structured morphotropic phase boundary (MPB) at room temperature [4, 5]. A survey of research studies on $Pb(Zr_{0.53}Ti_{0.47})O_3$ ceramic shows that a systematic study of the effects of antimony (Sb) about the properties not been reported [6, 7]. There is also no reporting about synthesis $Pb(Zr_{0.53}Ti_{0.47})O_3$ ceramics addition of Sb on the A site of Perovskite structure by solid state reaction (sintering temperature = 1423 K) which is the study of this paper.

Materials and Methods

The method synthesized the piezoelectric ceramics $(Pb_{1-x}Sb_x)$ $(Zr_{0.53}Ti_{0.47})$ O_3 (PSbZT) when x = 0.0, 4.5, 6.0, 7.5 mol% substrate of starting from oxide compound (PbO, Sb_2O_3, TiO_2, ZrO_2), weighing the stoichiometry of the substrate which has already calculated. Mixing precursors in deionized water with a ball – milled for 24 h, then the pelletizing with a hydraulic uniaxial machine with force 5,000 kPa and calcined at 1123 K for 4 h. After that take PSbZT powdered it amounted to 1 gram mixed with PVA, and then pelletizing with a hydraulic uniaxial machine with force 5,500 kPa before the sintered.

The PSbZT ceramic were sintered at 1423 K for 2 h in and prevents the evaporation of PbO_2 with the buried in PZT (53/47) powder while sintering. When PSbZT ceramics through sintering then sanded with sandpaper to crystal structure analysis by X – ray diffraction (XRD, SHIMADZU 6000, Japan) and calculate crystal size by Scherer equation, measured density by

Density Kit (MS204, METTLER TOLEDO, Switzerland) and Vickers hardness by Micro Vickers Hardness Tester (HMV-2, SHIMADZU) then coat silver glue to measured dielectric property and ferroelectric property.

Results and Discussion

The crystal structure of $(Pb_{1-x}Sb_x)$ $(Zr_{0.53}Ti_{0.47})$ O_3 (PSbZT) ceramics when x = 0.0, 4.5, 6.0 and 7.5 mol% was determined by XRD shown in Figure 1. The peaks appearing in the XRD traces of these are attributed to Sb were the increase in intensity can be evident by increasing of Sb volumes content [8 – 9]. This is evident in the planes (002), (200) and (112), (211) make aware of the crystal structure changes from tetragonal to be rhombohedral and lattice parameter a = b = 4.041 Å, c = 4.147 Å. This can be confirmed from PDF # 00–033–0784. The crystal size is 267.29 Å, 292.11 Å, 686.05 Å and 642.89 Å when x = 0.0, 4.5, 6.0 and 7.5 mol% respectively. The ratio of Sb x = 0.0 mol% and x = 4.5 mol% is a minimum crystal size. The ratio of Sb x = 6.0 mol% is a maximum crystal size increased when x = 6.0 mol% and x = 7.5 mol% as a result of changing the crystal structure of the PSbZT ceramics. The density of PSbZT ceramics is 7.51 g cm^{-3}, 7.41 g cm^{-3}, 7.22 g cm^{-3} and 7.45 g cm^{-3}. The Vickers hardness of PSbZT ceramics is 425.40 Nmm^{-2}, 496.20 Nmm^{-2}, 572.20 Nmm^{-2} and 614.60 Nmm^{-2} when x = 0.0, 4.5, 6.0 and 7.5 mol % respectively. When increasing the amount of Sb resulting in density and hardness of PSbZT ceramic decrease this is due to the crystal structure changes. The lattice parameter of PSbZT ceramics when x = 6.0 mol% is 0.9741 Å which the value is different from other ratios of Sb as a result of crystal structure changing of PSbZT ceramics. The overlap between crystal structure tetragonal and rhombohedral as shown in table 1.

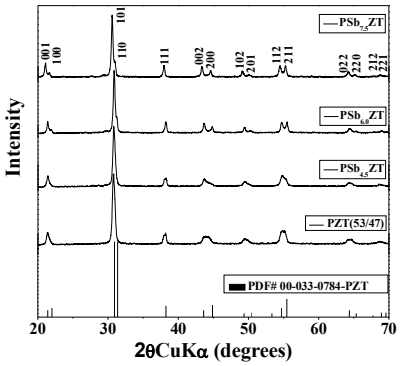

Fig. 1 XRD pattern of PSbZT ceramics were sintered at 1,423 K for 2 h

Table 1 Show experiment density, theoretical density, relative density and lattice parameter of PSbZT ceramics.

Sample	Density (g cm^{-3})	Theoretical density (g cm^{-3})	Relative density (%)	Lattice parameter (Å)		
				a	c	a/c
PZT	7.51	7.85	95.80	4.041	4.147	0.9742
$PSb_{4.5}ZT$	7.41	7.89	93.90	4.027	4.150	0.9704
$PSb_{6.0}ZT$	7.22	7.48	96.60	4.041	4.148	0.9741
$PSb_{7.5}ZT$	7.45	7.83	95.10	4.043	4.150	0.9742

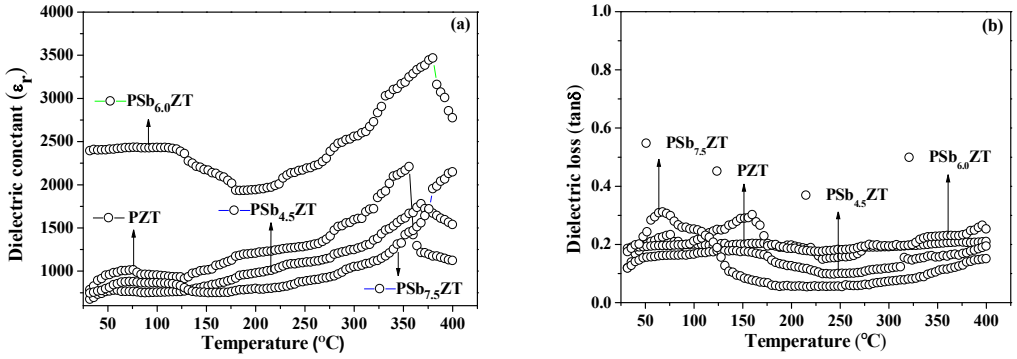

Fig. 2 The relative dielectric constant (a) and dielectric loss (b) at the function of temperature and frequency 1 kHz. of PSbZT ceramics.

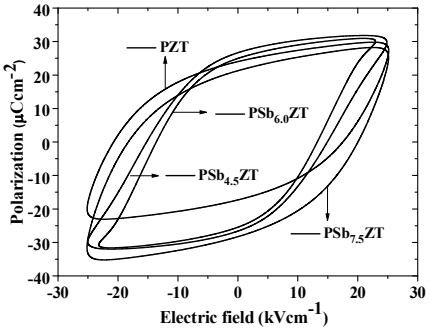

Fig. 3 Room temperature P – E hysteresis loops of the PSbZT ceramics at electric field of 25 $kVcm^{-1}$ and the frequency of 100 Hz.

The dielectric constant and dielectric loss of PSbZT ceramics show in Fig. 2. When measuring the temperature of 305 – 673 K at a frequency of 1 Hz. Found that when the adding Sb in ratio 6.0 mol% the maximum dielectric constant which is 3,464.46 at temperature 653 K and the frequency of 1 Hz. This shows that when Sb is added, the result is dielectric constant the increase compared to PZT (53/47) which is 2,206.78 at temperature 629 K and the frequency of 1 Hz. The dielectric loss show in figure 2 (b) it can be seen that when the adding Sb in ratio 6.0 mol% at 433 K and frequency 1 Hz. which is 0.302, compared to PZT (53/47) ceramic at the same temperature found that PZT (53/47) ceramic which is 0.203. As a result, it is confirmed that the adding Sb effect to behavior of dielectric constant and dielectric loss of PSbZT ceramics. It is also predicted that polar relaxation connected to the impurities and motion of the ionic particle's domain wall will cause dependence on frequency dependence on the insulating properties of the PSbZT ceramics [10]. The polarization versus electric field (P – E) hysteresis loops of PSbZT ceramics under different compression stresses during loading is shown in Fig. 3. Obviously, the shape of the hysteresis loop is different this is a result of the amount of Sb. It can be seen that the remanent polarization (P_r) increased by the ratio of Sb added to PZT (53/47) except that Sb is x = 7.5 mol% lowest drop. This confirms that well the behavior of PZT (53/47) ceramic when adding Sb it has changed in a good way, say that when adding Sb at the A site of PZT (53/47) ceramics can improve dielectric and ferroelectric properties, but must be added to the right amount. Therefore from this experiment the right amount for adding Sb at the A site of PZT (53/47) ceramics is x = 6.0 mol%.

Conclusion

PSbZT ceramics at x = 0.0, 4.5, 6.0, 7.5 mol% were synthesized by solid state reaction method. The crystalline structure obtained by XRD analysis was found to be a Perovskite structure there is an overlap between the crystal structure tetragonal and rhombohedral, which is a crystal structure morphotropic phase boundaries (MPB) [11]. The density and Vickers hardness decreased by the amount of Sb with the least at x = 6.0 mol%. It can be seen that dielectric and ferroelectric properties the changes are likely to improve from addition Sb. The effect of the addition Sb at the A site of PZT (53/47) ceramics points out that a good way to improve the dielectric and ferroelectric properties of PZT ceramics.

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