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Effect of Poling Parameter on Piezoelectric and Dielectric Properties of La and Sr Doped PZT

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ABSTRACT. La and Sr co-doped, modified lead zirconium titanate (PZT) ceramics, with composition $Pb_{0.93}La_{0.02}Sr_{0.05}(Zr_{0.52}Ti_{0.48})O_3$ was synthesized by dry-mixed planetary mill. The incorporation of La^{3+} and Sr^{2+} in the PZT lattice enhanced the tetragonality and suppressed grain growth thereby promoted piezoelectric and dielectric properties of the ceramics. The objective of this research is to determine the optimum poling parameters on La and Sr co-doped PZT (PLSZT) for better electrical properties. The optimum poling parameters (temperature, electric field and time) have been investigated via design of experiment (DOE) concept for the effective results. The results showed that when using the poling temperature at 140 °C with 4 kV/mm of electric field, the d_{33} and ε_r values were found to be highest at 327 pC/N and 1620, respectively. It revealed that poling temperature and applied electric field plays a dominant role in influencing electrical performance of PLSZT which may be suitable for possible device applications.

Keywords: *PZT, Doped PZT, Poling, DOE, Piezoelectric;*

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1. INTRODUCTION

Recent advances in ceramics have provided greater control over aspects of composition and microstructure that govern physical properties. Such control makes it possible to tailor ceramics with special chemical, thermal, mechanical, and electrical requirements [1]. Lead zirconate titanate (PZT) is one of the advance ceramics which has great technological interest due to their excellent piezoelectric and ferroelectric properties. It is also known as a ceramic perovskite material and it shows a marked piezoelectric effect widely used for in the microelectronics industry. The piezoelectric material is the materials that can create electricity when subjected to a mechanical stress also known as direct piezoelectric effect. They will also work in reverse, which is generating a strain by the application of an electric field and is known as indirect piezoelectric effect [2]. Many base of PZT composition have been manufactured by chemical modification in order to satisfy the requirements and enhance the PZT properties especially in electrical properties.

In order to improve the electrical properties, many investigations have been done on altering the composition of the piezoelectric materials, which is by doping with other materials. Besides, the poling parameters also give the effect towards the electrical and mechanical properties of the system especially in dielectric and piezoelectric properties. The previous researchers had investigated La and Sr co-doped PZT to improve the dielectric and piezoelectric properties [3]. Some researchers also have been study the poling effect of PZT ceramics but not many reports on doped PZT samples [4,5]. Based on Li et al. it is generally believed that the temperature plays an important role during the poling process because it leads to the best piezoelectric properties.

However, there were still remains a need for an efficient method that can achieve optimum poling parameters for doped PZT which is by using statistical design method. Therefore, this research came out with the objective to determine the optimum poling parameters on La and Sr co-doped PZT (PLSZT) based system using Design of Experiment (DOE) software. DOE is used because it is a design experiment which is a methodical way of intentionally changing a process and give the effective results [6].

2. MATERIALS AND METHODS

PbO, ZrO_2, TiO_2, La_2O_3 and $SrCO_3$ powders were used as starting materials to synthesize the La- and Sr-doped PZT. The mixture powders were prepared according to the stoichiometric ratios of $Pb_{0.93}La_{0.02}Sr_{0.05}(Zr_{0.52}Ti_{0.48})O_3$, then the mixture was dry-mixed in a planetary mill for 40 hours. The milled powder was then pressed into pellets with 13 mm diameter with compaction pressure at 200 MPa and sintered at 1200 ºC for 3 hours and then was undergo the characterization.

Phase formation of sintered samples were characterized using X-ray diffraction (XRD) technique. Meanwhile, the pellet samples were polished and thermally etched at 950 °C for microstructure analysis by using field emission scanning electron microscopy (FESEM) technique. For poling process, the pellet samples were polished and the both surface of samples were coated with silver paste. Then, the samples undergoes poling process by applying the surface sample in a contact with different electric fields (2, 3, and 4 kV/mm), temperature (100, 120 and 140 °C) and times (10, 15 and 20 min).

2.1 Design of Experiment (DOE)

DOE was used for La and Sr co-doped samples (PLSZT) after sintering. The effect of variables, such as temperature (X_1), electric field (X_2) and time (X_3) were studied using response surface methodology (RSM) based on central composite design (CCD). Factors used were 2^3 with 3 centre points and α value was 1.682. Table 1 shows the experimental conditions of this work.

Table 1 Control factors and their levels in poling parameters using RSM

Parameter	Code	Level				
		Low, -1	Medium, 0	High, +1	Star point, α	
					-α	+α
Temperature (°C)	X_1	100.0	120.0	140.0	86.5	153.5
Electric field (kV/mm)	X_2	2.0	3.0	4.0	1.3	4.7
Time (min)	X_3	10.00	15.00	20.00	23.41	7.00

The sequence of poling parameters was carried out according to design of experiment (DOE) set by Minitab 16 Software (Response surface methodology by central composite design). Table 2 displays the sequence of poling parameters.

3. RESULTS AND DISCUSSION

3.1 Phase Analysis.

Fig. 1 shows the XRD patterns of sintered PZT and PLSZT samples. Both diffractograms can be fully-indexed to a single-phase tetragonal perovskite structure with a space group of P4mm. Besides, the peaks were shifted to the higher angle after addition of dopant materials due to the slight variation in lattice constant which indicates the substitution of La ion in the crystal lattice. When the peaks shifted to the higher angle, the lattice parameters contracted dueto the difference in ionic radii between the Pb element (119 pm) and the La and Sr co-dopant ions (103 and 118 pm respectively) with creation of oxygen vacancy.

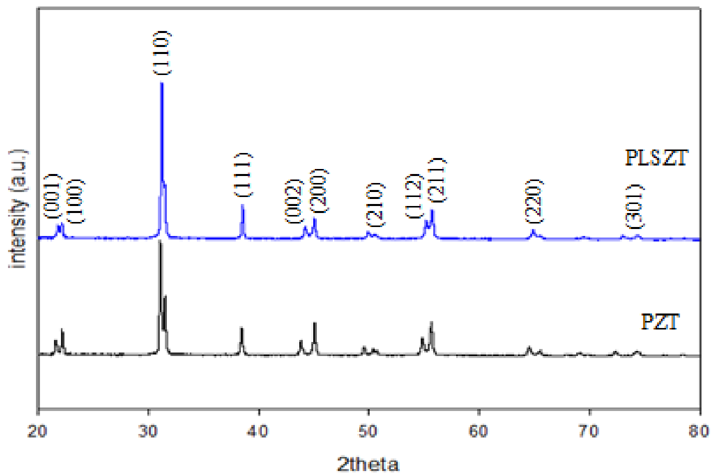


Fig. 1 XRD pattern of sintered pellet of pure PZT and PLSZT

Table 2 DOE of poling parameters using RSM

Std. Order	Run Order	Temperature (°C)	Electric field (kV/mm)	Time (min)
12	1	120.0	4.7	15.00
4	2	140.0	4.0	10.00
13	3	120.0	3.0	7.00
8	4	140.0	4.0	20.00
7	5	100.0	4.0	20.00
2	6	140.0	2.0	10.00
16	7	120.0	3.0	15.00
6	8	140.0	2.0	20.00
9	9	86.5	3.0	15.00
1	10	100.0	2.0	10. 00
15	11	120.0	3.0	15.00
17	12	120.0	3.0	15.00
5	13	100.0	2.0	20.00
14	14	120.0	3.0	23.41
3	15	100.0	4.0	10.00
11	16	120.0	1.3	15.00
10	17	153.5	3.0	15.00

The peaks shift to the higher angle as the doping La due to substitution of La^{3+} (1.22 Å) and Sr^{2+} (1.27 Å) to Pb^{2+} (1.32 Å). Due to slight variation of atomic radius, strain will be generated in the matrix and cause the reduction in crystallite size in PLSZT. The peak intensity was also increased after adding the dopant due to the increasing of the number of oriented crystallite. Same observation also was made by Law et al. [7] which found that the increase of the peak intensities on XRD pattern was due to high number of domains oriented in the sample.

3.2 Microstructure Analysis.

Fig. 2 shows FESEM micrographs for undoped PZT and PLSZT samples. The micrographs show the grain sizes of PLSZT was homogeneously distributed throughout the sample with certain degree of porosity. The grain size was decreased with addition of dopants in PZT system (Fig. 2b). The reduction of grain size was due to the small substitution of La and Sr into Pb site and the others are precipitated on grain boundary and then suppressed the grain growth. This is because the crystal's interior must be presented in lot of vacancy (dopedcations and oxygen) or there is no substitution has been occurred. Therefore, the deficiency of Pb led to increase vacancies at Pb and O sites, and subsequently, reducing the grain size. Bahanurddin et al. [3] also claimed that addition of La in PSZT shows the grain growth has been slowed down.

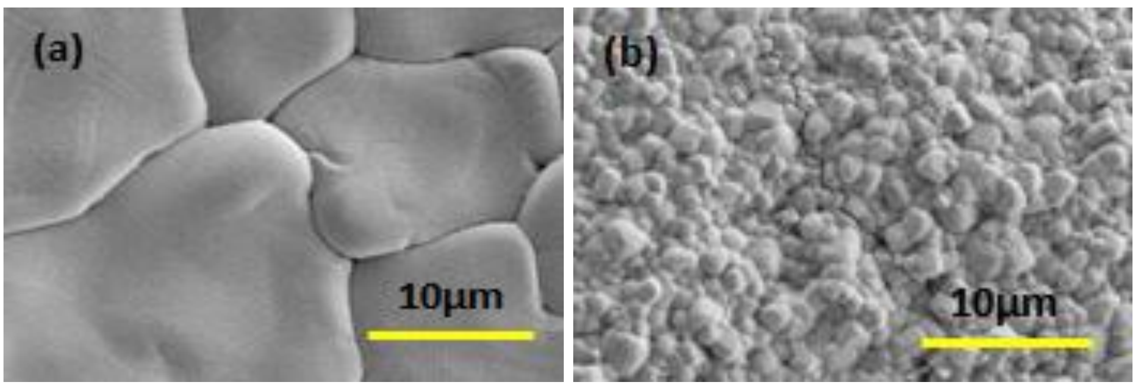


Fig. 2 Surface view for (a) PZT and (b) PLSZT at 1000x magnification

3.3 Piezoelectric coefficient.

Based on DOE result, the identifications of significant factor and interaction effect were made using analysis of variance (ANOVA). The quadratic model relationship was significant at $\alpha = 0.05$. In addition for lack of fit, P >0.05 point out the model is in good fit. Table 3 tabulated the ANOVA for this work.

From the ANOVA results, a mathematical model in Eq. 1 can be expressed in order to predict piezoelectric effect in poling process with various poling parameter. This equation was formed based on the regression coefficient of piezoelectric coefficient. The standard deviation, R^2 and R^2 adjusted were 98.08% and 95.61% respectively. These indicate that the accuracy of model is 98.08%. This entire statistical test showed that the develop model was suitable for representing data and provide good relationship between variables and response.

$$Y = 75.256 + 1.611X_1 + 68.861X_2 + 1.3X_3 - 0.004X_1^2 - 6.304X_2^2 - 0.04X_3^2 - 0.138X_1X_2 + 0.003X_1X_3 - 0.05X_2X_3 \quad (1)$$

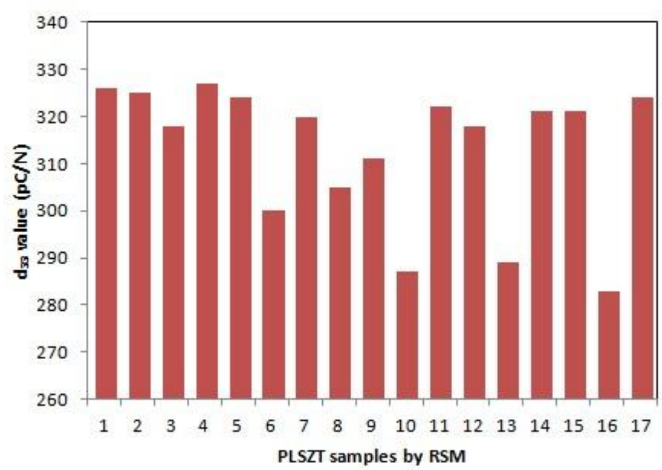
$Y = d_{33}$
X_1 = Temperature
X_2 = Electric field

X_3=TimeFactors of poling parameters point out positive values respectively. Increasing these factors will raise d_{33} value. In conjunction, the equation also indicates d_{33} more influence by electric field than temperature and time.

Fig. 3 shows piezoelectric coefficient (d_{33}) of PLSZT based on DOE run order. The value shows the increasing the temperature and electric field increased the d_{33} value (140 °C, 4 kV/mm and 20 min). However, there was not much different in d_{33} between others. Prewitt and Jones [5] also claimed by increasing of electric field and temperature increased the d_{33}. This is because at an elevated temperature, higher degree of domain reorientation developed larger polarization in ceramic. Thus, it contributed to the higher the d_{33}. At high electric field, the absolute extrinsic contribution due to irreversible displacement of domain walls increase in samples more highly poled.

Table 3 Analysis of variance ANOVA for piezoelectric properties

Source	Hypothesis, P	Regression coefficient
Constant	-	75.256
Linear:		
Temperature	0.001	1.611
Electric field	0.000	68.861
Time	0.177	1.300
Square:		
Temp x Temp	0.105	-0.004
Electric field x Electric field	0.000	-6.304
Time x Time	0.311	-0.040
Interaction:		
Temp x Electric field	0.039	-0.138
Temp x Time	0.825	0.003
Electric field x Time	0.825	-0.050
Lack of Fit	0.275	

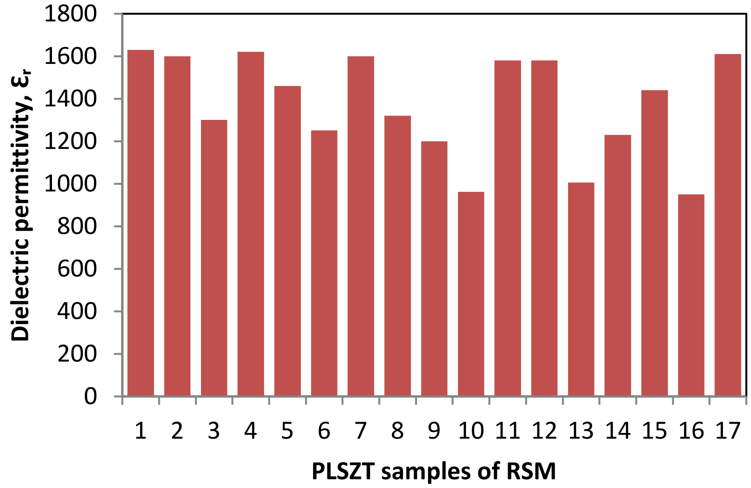


Run Order	Temperature (°C)	Electric field (kV/mm)	Time (min)
1	120.0	4.7	15.00
2	140.0	4.0	10.00
3	120.0	3.0	7.00
4	140.0	4.0	20.00
5	100.0	4.0	20.00
6	140.0	2.0	10.00
7	120.0	3.0	15.00
8	140.0	2.0	20.00
9	86.5	3.0	15.00
10	100.0	2.0	10. 00
11	120.0	3.0	15.00
12	120.0	3.0	15.00
13	100.0	2.0	20.00
14	120.0	3.0	23.41
15	100.0	4.0	10.00
16	120.0	1.3	15.00
17	153.5	3.0	15.00

Fig. 3 d_{33} values of PLSZT samples corresponding to RSM

3.4 Dielectric permittivity.

Fig. 4 shows the dielectric permittivity (ε_r) of all the PLSZT samples at 1 MHz. Among all of the samples, the sample number 4 had the highest value of ε_r (1620) while the sample number 16 exhibits lowest ε_r (950). Sample 4 was poled at high temperature (140 °C) while sample 16 was poled at 120 °C. It shows the highest ε_r was obtained at higher poling temperature because of the increasing in domain wall motion during poling process. Meanwhile, at low temperature the movement of the domain was slow and the dipole was not fully aligned and produced a low dielectric permittivity.



Run Order	Temperature (°C)	Electric field (kV/mm)	Time (min)
1	120.0	4.7	15.00
2	140.0	4.0	10.00
3	120.0	3.0	7.00
4	140.0	4.0	20.00
5	100.0	4.0	20.00
6	140.0	2.0	10.00
7	120.0	3.0	15.00
8	140.0	2.0	20.00
9	86.5	3.0	15.00
10	100.0	2.0	10. 00
11	120.0	3.0	15.00
12	120.0	3.0	15.00
13	100.0	2.0	20.00
14	120.0	3.0	23.41
15	100.0	4.0	10.00
16	120.0	1.3	15.00
17	153.5	3.0	15.00

Fig. 4 Dielectric permittivity of PLSZT samples at different poling parameter

4. SUMMARY

La and Sr co-doped PZT ceramics were produced via high planetary mill. The perovskite structure was exists in PLSZT. The addition of dopants decreased the grain size of sample and increased the d_{33} value. Meanwhile, the result of poling parameters based on DOE shows the highest value of d_{33} (327 pC/N) and ε_r (1620) at temperature (140 °C) and electric field (4 kV/mm).

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