Fabrication and Characterization of Li_2CO_3 Modified Low Temperature Sintered PZT-5H Compositions for Multilayer Piezoelectric Ceramics

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Abstract In the use of piezoceramic applications, piezoceramic compositions have high d_{33} (>500 pC/N) and K_t (2000) such as PZT-5H. However, these ceramics can be sintered at high temperatures up to 1200°C . Since these require the use of Ag-Pd inner electrodes with higher Pd ratio, multilayer ceramics are not preferred due to their high costs. In this study, it was aimed to reduce the sintering temperature without dramatically losing the electrical properties of the PZT-5H ceramics with high d_{33} , k_p and dielectric constant values. In this context, the PZT-5H composition was modified with different proportions of Li_2CO_3 and sintered for 2-4 hours at different sintering temperatures such as 875°C - 1250°C depending on the ratio of additions. The piezoelectric and ferroelectric properties of the ceramics were investigated and obtained values were compared with undoped PZT-5H. It was observed that the d_{33} values of the different amount of Li_2CO_3 added compositions sintered at optimum sintering temperature are close that the composition without additives. In this way, it is predicted that 9/1 Ag/Pd and Ag inner electrode materials can be used in multilayer production and their costs can be reduced.

Keywords Multilayer, PZT-5H, Low Sintering, Li_2CO_3

1. Introduction

Piezoelectric ceramics convert mechanical energy into electrical potential and vice versa. For this sensing and actuating performance, piezoelectric materials are used in many sectors from aviation to defence, automotive to medical applications. Depending on the application area, new needs and performance have emerged in the use of bulk piezoelectric ceramics. Therefore, studies have been carried out on structures of multilayer piezoelectric materials. Multilayer piezoelectric ceramics are used for low driving voltage, high generative force and stability under loading [1]. Sintering is the most important step in the production of piezoelectric ceramics that have high piezoelectric properties and it depends on composition. Sintering temperatures of PZT-based materials are in the range of 1100°C - 1300°C [2]. One of the most important constraints in the production of multilayer ceramics is that the inner electrodes that should be used expensive un-reactive electrodes like Pt [3] or Ag-Pd inner electrodes [4] with high Pd ratio due to high sintering temperatures. In this case, the production costs of multilayer ceramics increase. It can be used Ag-Pd inner electrodes with a lower Pd ratio to reduce production costs [5]. However, it is known that thermal mismatch is more likely to occur when using these electrodes under these

conditions [6]. By reducing the sintering temperature of PZT-based ceramics, it is possible to produce multilayer ceramics by using less costly Ag-Pd electrodes that have lower Pd ratio or Ag inner electrode for different applications [7]. Various studies were carried out to reduce sintering temperatures of piezoelectric ceramics. One of them is increasing the grinding time either after calcination or reducing the initial raw materials particle size [8-12]. The other method to reduce sintering temperatures is using sintering agents. Li_2CO_3 [13], ZnO, CuO [14], CuO [15,16], PZN in PZT [17], $\text{x}\text{Bi}_2\text{O}_3\text{-yBi}_2\text{O}_3\text{-zCdO}$ frit for PZT [18] La_2O_3 [19], $\text{CaO-Fe}_2\text{O}_3$ [20] can be used to reduce sintering temperature. The added Li_2CO_3 passes into liquid phase and provides more dense structure. There are some studies in the literature lowering sintering temperature of PZT-based compositions. Li_2CO_3 addition is used in PZN-PMN-PZT composition systems and decreased the sintering temperature. [21]. The effect of Li_2CO_3 additive on the decrease of sintering temperature, microstructural and electrical properties in PMN-PZT based systems was investigated [22]. However in the study, the piezoelectric properties were partially examined and no information was given about the ferroelectric properties. Its usability in PMN-PFN-PZT composition systems also has been investigated and multilayer ceramics were produced by using Ag inner electrode [23]. Although some works have been encountered in the literature that Li_2CO_3 addition provides decrease on the sintering temperature of commercial PZT composition, there are some shortcomings about piezoelectric and ferroelectric properties to be completed [24].

In this study, sintering temperature was decreased by adding Li_2CO_3 to PMN-PZT powder which can be sintered at high temperatures and piezoelectric and ferroelectric properties of these ceramics were investigated. In our study, a systematic approach was carried out about decreasing sintering temperature of PZT composition, it was observed that the sintering temperature decreased without dramatic deterioration on the electrical properties. Thus, PZT-5H ceramics that can be sintered at lower temperatures were produced. In this way, multilayer ceramics that can be sintered at lower costs by using the Ag-Pd inner electrode with a lower Pd ratio. This study is the preliminary composition research of the multilayer ceramic production that will be produced using low cost Ag inner electrode which will be carried out later. Aim of this study is the sintering the PZT-5H ceramics at 950°C - 1000°C and 850°C - 900°C by obtaining

dense and homogenous microstructure without deteriorating the electrical properties for two different (9/1 and 10/0) Ag-Pd ratio multilayer inner electrodes. For this reason, determining optimum amount of Li_2CO_3 addition at optimum sintering temperature is essential.

2. Materials and Methods

PZT-5H composition (SunnyTech Piezo) has been modified with different amounts ($x=0.0, 0.05, 0.075, 0.1, 0.2\text{wt}\%$) of Li_2CO_3 powder. PZT-5H composition is pressed and sintered at 1250°C for 2 hours. For Li_2CO_3 added compositions, first binder removal process of PZT-5H Powder was evaluated at 650°C -2 hours. PZT-5H and Li_2CO_3 powder was ball milled about 24 hours in alcohol medium. Then PVA added as a binder to dried powder composition and granulated. Obtained powder was pressed into pellets 12,7 mm diameter and 1,4 mm thickness under 200 MPa and sintered at temperature from 875°C to 1275°C for 4 hours in alumina crucible depending on the Li_2CO_3 content.

Crystal structure of the materials was determined by using XRD. The densities were measured with Archimedes Method. The microstructures of fractured sample were analyzed by scanning electron microscope (SEM, Type: Zeiss EVO). The piezoelectric coefficient d_{33} values were measured by using APC Pennebaker d_{33} meter. The piezoelectric parameters of resonance frequency, impedance and capacitance were measured by using Agilent 4294A impedance analyzer, Q_m , k_p , and tangent loss ($\tan \delta$) were calculated.

3. Results and Discussions

Figure 1 shows the density of the different amount of Li_2CO_3 modified compositions of PZT depending on the sintering temperature. PZT-5H composition has $7,75 \text{ g/cm}^3$ density value at 1250°C sintered for 2 hours. Desired densification occurs above 1100°C for PZT samples without sintering agent Li_2CO_3 .

$0.05\text{wt}\%$ and $0.1 \text{ wt}\%$ Li_2CO_3 added compositions exhibit $7,75 \text{ g/cm}^3$ at 950°C , $0.2\text{wt}\%$ Li_2CO_3 added composition exhibits same density value at 850°C sintering temperature. It can be seen that the added amount of Li_2CO_3 has been increased, sintering temperature is decreasing. It can be seen that Li_2CO_3 act as sintering agent.

Figure 1. Density of the Li_2CO_3 added PZT compositions depending on sintering temperature

Figure 2 shows the XRD patterns of compositions with various Li_2CO_3 doping content depending on optimum sintering temperature.

Figure 2. XRD patterns of Li_2CO_3 added compositions a) 0.2wt% Li_2CO_3 added compositions depending on sintering temperature b) different amount of Li_2CO_3 added compositions that sintered at optimum sintering temperatures.

All samples have perovskite phase without any secondary phase. Tetragonality increases when Li_2CO_3 is added to the PZT-5H composition. Lithium ions replace the B-sited Zr^{4+} ($0.72\text{\AA}$) and Ti^{4+} ($0.61\text{\AA}$) atoms and occupy the octahedral sites of ABO_3 lattice depending on their small atomic radius ($0.74\text{\AA}$). Distortion occurs in the lattice that c axis is lengthened [21].

b

Figure 3. Microstructure of PZT-5H composition depending on sintering temperature a) 1250°C-2h b) 975°C-2h

It can be seen from the figure 3a, uniform and dense microstructure has been obtained at 1250°C for PZT-5H specimen. In figure b, image relates to PZT-5H sample has been sintered at 975°C for 2h. Porous structure can be seen, grains can not be occurred in the structure, this means sample can not be sintered.

a

b

c

Figure 4. Microstructure of %0.05 Li_2CO_3 doped composition depending on sintering temperature a) 900°C b) 950°C c) 975 °C

a

b

c

Figure 5. Microstructure of % 0.1 Li_2CO_3 doped composition depending on sintering temperature a) 900°C b) 950°C c) 975 °C

a

b

c

Figure 6. Microstructure of % 0.2 Li_2CO_3 doped composition depending on sintering temperature a) 825°C b) 850 °C c) 875 °C

Figure 4 shows the results of the fractured surface of Li_2CO_3 added PZT-5H ceramic sample bodies with increasing temperature that is close to optimum sintering conditions for increasing amount of Li_2CO_3 . In Figure 4 b it can be seen that liquid phase occurs around the particles

for 0.05 wt% Li_2CO_3 added sample sintered at 950°C. Although final microstructure of sample has been occurred, porous structure can be seen. Dense and homogenous microstructure can not be seen in 0.05 wt% Li_2CO_3 added to PZT-5H at 950°C-975°C. When the

amount of Li_2CO_3 has increased to 0.1wt% and sintered at 900°C (figure 5a), grains occur but grain growth can be seen in figure 5b while increasing temperature. Excess Li ions unable to settle in the perovskite lattice can cause abnormal grain growth by pinning at grain boundaries. However Li ions precipitate at grain boundaries and prevent grain growth [22]. At 975°C sintered sample (figure 5c), dense and homogenous microstructure can be obtained. Transient liquid phase was formed by Li_2CO_3 in sintering stages [26]. Li_2CO_3 has low melting temperature (723°C) that forms liquid phase at the initial and intermediate stages of sintering process. Liquid phase wets and covers the surface of grains and promote the capillary drag for densification. In the last stage of sintering, Li atoms are cited into the B-site of lattice and modification of perovskite occurs [25]. From the results, it is obvious that Li_2CO_3 addition enhances the densification of sintered bodies of PZT-5H ceramics at lower temperatures that need undoped form of composition. Temperature is a parameter that affects driving force to increase the grain size [27], therefore sintering temperature of lithium doped PZT-5H was kept around 875°C-975°C proportional to Li content of compositions much lower than 1200°C-1300°C that of undoped PZT-5H ceramics. When the added amount of Li_2CO_3 was

increasing to 0.2 wt%, sintering temperature was decreasing to 875°C to obtain desired microstructure. Liquid phase residues were apparent at 850°C.

The piezoelectric properties of the pellets sintered at different temperatures using modified and unmodified compositions are given in table 1.

Undoped PZT composition has 600 pC/N d_{33} , k_t of 0.65 values sintered at 1250°C for 2 hours. 0.05 wt% Li_2CO_3 added composition has 630 pC/N d_{33} and 0.62 k_t value by sintering at 975°C for 4 hours, 0.2 wt% Li_2CO_3 added composition also achieves that values 875°C for 4 hours. Dielectric constant values remained relatively low in 0.05 and 0.1 wt% Li added compositions. As the added Lithium amount and sintering temperature increase, the porosity values decrease, thus the dielectric constant values increase [28]. Li^+ atoms substitute B site atoms of ABO_3 that have 4+ valence mentioned before and cause oxygen vacancies. Although this distortion causes clamping effect of domain walls and hard poling of Li doped ceramics, grain size controlled by sintering temperature takes part in the literature [25]. As can be seen from the table, the desired piezoelectric properties and sintering temperatures from the PZT-5H composition were achieved by adding appropriate amount of Li_2CO_3 .

Table 1. Piezoelectric properties of modified and unmodified compositions sintered at different temperatures

Li_2CO_3 wt%	Temperature	d_{33} (pC/N)	Q_E	Dissipation Factor	k_t	Q_m	K^T
0.0	1225 °C	569	61	0,0163	0,68	61	4241
0.0	1250 °C	600	58	0,0172	0,65	66	3881
0.05	900 °C	388	41	0,0241	0,37	77	2639
	950 °C	597	56	0,0175	0,55	79	3091
	975 °C	630	52	0,0192	0,62	75	3546
0,1	900 °C	500	39	0,0256	0,49	57	3073
	950 °C	540	55	0,0181	0,58	65	3218
	975 °C	580	53	0,0188	0,60	64	3435
0,2	825 °C	624	37	0,0270	0,54	51	4102
	850 °C	673	38	0,0256	0,61	29	3879
	875 °C	710	42	0,0238	0,64	49	3990

Figure 7 shows the ferroelectric hysteresis loops for different Li_2CO_3 amount added compositions depending on sintering temperature.

Figure 7. Hysteresis loops of different compositions depending on sintering temperature a) 0.2 wt% Li_2CO_3 added composition b) 0.1 wt% added composition c) 0.05 wt% added composition d) comparison of Li_2CO_3 doped compositions at optimum sintering temperatures

Polarization measurements depending on applied electrical field (P-E) graphs are given in figure 7. In all Li_2CO_3 doped compositions, remnant polarization values increase as the sintering temperature increases. The coercive electrical field values are similar for the materials added 0.1 wt% and 0.2 wt% Li_2CO_3 compositions. In the

0.05wt% Li_2CO_3 added composition, the squareness of the hysteresis loop seems very low since the sintering temperature is about 900°C is too low for sintering. As the sintering temperature of the lithium added compositions increased, the remnant polarization values increased. It is inferred that lower remnant and saturated polarization

values correspond to undensified microstructure of the material [26]. In figure 7d, All compositions that sintered at optimum sintering temperatures were compared, remnant polarization values were between 28-30 $\mu\text{C}/\text{cm}^2$ and saturated polarization values have small differences. These small differences are related to domain wall movements and clamping effects [25].

Strain versus electrical field graphs can be seen in figure 8. As the sintering temperatures increase and reach the optimum temperature, symmetry is observed in the strain curves related to dense microstructure mentioned above. In figure 8d, Li_2CO_3 added PZT samples has

higher strain values than undoped sample, due to extrinsic effects of domain wall contributions [29]. Lithium atoms settle at grain boundaries and domain boundaries, increasing the disorder. Therefore, neighbour domains movement increase the strain value by causing the clamping effect. Li_2CO_3 added compositions have desired ferroelectric and piezoelectric properties corresponding to large and low field measurements. For this reason, compositions with 0.1wt% and 0.2 wt% Li_2CO_3 added compositions that have lower sintering temperatures than 1000°C , can be used instead of the PZT-5H composition.

Figure 8. Butterfly curves of different compositions depending on sintering temperature a) 0.2 wt% Li_2CO_3 added composition b) 0.1 wt% added composition c) 0.05 wt% added composition d) comparison of Li_2CO_3 doped compositions at optimum sintering temperatures

4. Conclusion

The effect of Li_2CO_3 addition on the sintering behaviour of commercial PZT-5H ceramics was investigated. Two different piezoceramic compositions were developed to have sintering temperature lower than 1000°C . Piezoelectric coefficients and properties were not decreased too much from the original values of high temperature sintered PZT-5H ceramics. d_{33} values are acceptable values of 580-710 pC/N. Coupling factor values are also higher than 0.55. Dielectric constant values are changing about %5 for the whole ceramics. Optimum

sintering temperature for %0.1 Li_2CO_3 containing composition was 950°C , and for 0.2 Li_2CO_3 is 875°C . All lithium containing compositions show pure perovskite phase.

The sintering temperatures of Li_2CO_3 doped PZT-5H ceramics are low enough for sintering multilayer ceramics that can be manufactured by using inner electrode has lower ratio of Ag/Pd electrode containing precious Pd metal or Ag electrodes. After this study, the usability of the produced compositions in multilayer ceramic systems by using different Ag/Pd ratio inner electrodes will be investigated.

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