INFLUENCE OF FORMING PRESSURE OF COMPACTED POWDERS ON DENSIFICATION OF SINTERED BODY

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1. INTRODUCTION

Piezoelectric ceramics are widely used as active elements for a wide variety of ultrasonic transducers due to their extraordinary property of converting the electrical energy into mechanical strain and *vice-versa* at a high yield. The great variety of application of piezoelectric ceramics implies the use of piezoceramic active elements of different shapes and sizes and of good quality. The performances of transducers depend intrinsically on the quality of the piezoceramic elements, i.e. on their physical, electrical and piezoelectric properties. Amongst them, the density represents an important parameter. It depends on the internal structure (grain sizes and size distribution, pores, sintering temperature and some other technological parameters) of ceramic body. Particularly dense and uniform ceramics are desired when high performance applications such as piezoelectric motors, actuators, biomedical sensors or high power transducers are required. To achieve this, care must be taken during processing of ceramics and each step of the process must be carefully monitored. Especially the powder homogeneity, shape and size distribution of the particles. In addition shape and geometry as well as pressure pressing and sintering temperature may affect the quality of the final sintered body. The present investigation reports on the results concerning the influence of the structure of initial powder, shape and geometry of the compacted powder as well as the initial forming pressure on the final density of the sintered ceramic bodies, of a lead titanate zirconate (PZT) type powder, currently used for the fabrication of different ceramic parts for piezoelectric ultrasonic transducers.

2. OBJECTIVES

The main objective of this study was to investigate: **the effect of shape and initial forming pressure of piezoceramic submicronic compacted powders on densification of the final sintered body.** Since the overwhelming majority of application of piezoceramic uses disc or cylinder shaped samples as sensing elements we carried out the experiments on such samples. The densification degree of sintered samples was studied as function of:

- Sintering temperature
- Shape of the samples (Diameter/thickness ratio)
- Position within the sample
- Forming pressure at compaction

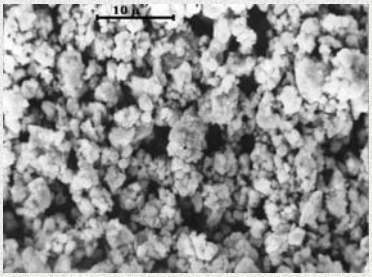

Figure 1
Photograph showing the grain size and shape of the PZT powder used in the experiment.

3. EXPERIMENTAL

The PZT powder was prepared by the mixed oxide route from p.a. purity oxides. The mixing was carried out in a planetary ball mill, in methanol, using agate vials and balls of 10 mm diameter and a ball/powder weighted ratio of 2:1. After mixing for 3 hours, the product was dried and double calcined at 850 °C and 900 °C followed by a final milling of 10 hours. This processing procedure produced a rather uniform submicron fine powder as can be seen in figure 1. Morphologic evaluation gave a specific surface area of about 1.6 m^2/g corresponding to an average spherical equivalent diameter of about 0.6 µm. We have carried out the pressing and sintering experiments on samples with geometries from cylinder to disc, having as working parameter the diameter to thickness ratios, D/h. The powder was compacted in cylindrical steel dies of different diameters, using uniaxial pressing and pressures from 40 to 180 MPa, thus covering the usual forming pressure for such materials. Pressures lower than 40 MPa were not able to produce consistent green samples while pressures higher than 180 MPa produced inconsistent friable samples. The densities of the green compacted powder were evaluated only geometrically while for the sintered ones both geometrically and Archimedes's method were used. The discrepancy between the two procedures was less than 1%, in some cases.

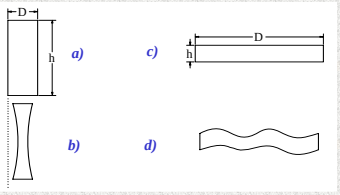

Figure 2
The change of shape after sintering for samples with very low and very high D/h ratio, respectively:
a) cylindrical sample before sintering;
b) cylindrical lenticular (biconcave) after sintering;
c) disc samples before sintering;
d) deformed disc after sintering

4. RESULTS

The sintering temperature interval for standard disc shaped samples of 20 mm diameter and 2 mm thickness, pressed at 100 MPa was between 1100 and 1350 °C. The density as a function of temperature is shown in figure 3. There is an optimum sintering temperature around 1250 °C, at which the densification reaches nearly 98% of theoretical density. Therefore, all sintering experiments were carried out at this temperature for a dwell time of 4 hours. Samples with D/h ratios between 2 and 16, were pressed at the lowest (60 MPa) and highest (180 MPa) pressures. Figure 4 illustrates the behavior of the density of sintered sample as a function of D/h ratio. There is an optimum D/h~10 ratio which provides samples with the highest density regardless the initial forming pressure. We have prepared samples with D=10 mm and h=40 mm (D/h~0.25) and D=40 mm and h=1.5 mm (D/h=26.7) respectively. After sintering their shapes changed and they looked, as it is exaggerately shown, like in figure 2. The density gradient of cylindrical samples both before and after sintering, was determined by slicing the cylinders in 20 thin pieces of about 1.5 mm thick along the long axis and numbered them from 1 (end cap) to 20 (the other end cap). The density of each slice is shown in figure 5. One may note the symmetric values of the density gradient. For the green compacted cylinder there is a density gradient of about 12 % between the ends and the middle while for the sintered sample practically there is none because the cylinder shrink much more at the middle where the green density is lower to compensate for the density differences, as expected, and as it was confirmed in figure 6 where the densities of the green compacted and sintered samples respectively are shown as a function of compacted pressure.

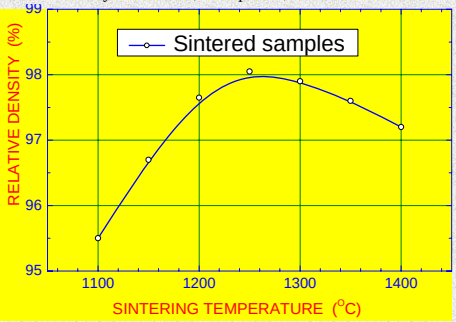

Figure 2
The dependence of the final density of sintered samples on the sintering temperature.

The density increases with increasing pressure, for the green compacted samples and remains practically constant for the sintered one regardless the forming pressure (Fig. 6). Such a behavior may be, at least qualitatively, assumed to be due to the differences between the friction coefficients of the die side wall and particles and interparticles respectively which may induce changes in the particles flow properties. It is interesting to remarque the independence of the density of sintered samples on the pressing pressure of green compacts due probably to a slight differences in the packing density of green compacted samples. The only visible effect of different initial packing densities could be expected for shrinkage (Fig. 7). Shrinkage continuously decreases with increasing forming pressure from 21 to about 17.5 % for diameter and from about 16 to 13 % for thickness respectively when pressure increases from 40 to 180 Mpa. For thinner discs (high D/h) the movement distances are lower in the axial direction and higher in the radial one, and consequently isles with different packing green densities may be formed which, in their turn, may produce sample deformation during the sintering process.

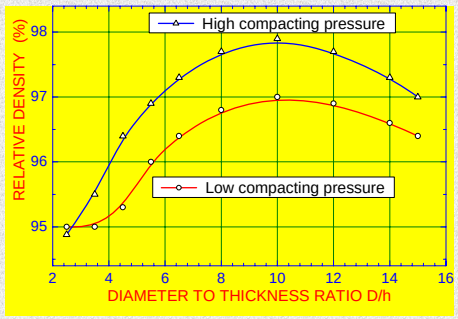

Figure 3
The dependence of the final relative density of sintered samples on the diameter to thickness ratio, for samples initially formed at high and low pressures.

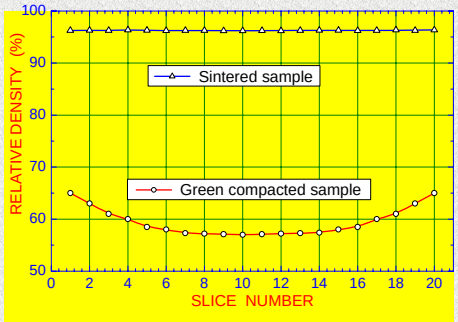

Figure 5
The dependence of the relative density for cylindrical green compacted and sintered samples, along the long axis of cylinder.

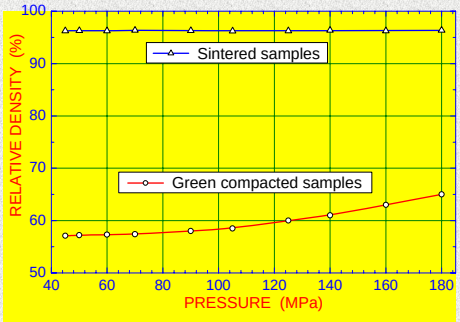

Figure 6
Green and sintered samples densities as a function of initial forming pressure.

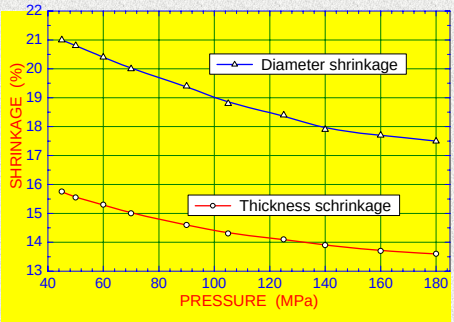

Figure 7
Diameter and thickness schrinkage of sintered disc samples as a function of the initial forming pressure

CONCLUSION

- Cylindrical shaped samples of a "soft" piezoelectric PZT type powder were compacted at pressures ranging from 40 to 180 MPa and then fired at 1250 °C for 4 hours.
- The densities of the sintered samples are practically independent on the initial forming pressure, i.e. on the green densities of the samples.
- The sample shrinkage decreased nearly linearly with increasing packing degree, more important being the shrinkage in diameter than in the thickness one.
- The density of the sintered samples slightly depends on the samples dimensions, the optimum value being reached for samples with a diameter to thickness ratio of about 10.
- All these findings are important technological factors very useful when mass production of piezoceramic parts is required.