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INFLUENCE OF PRESSING PRESSURE ON MICROSTRUCTURE AND ELECTRICAL CHARACTERISTICS OF $BaTiO_3$ CERAMICS

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ABSTRACT

The effects of pressing pressure on the microstructural characteristics and on electrical properties of pure barium titanate ceramic materials were investigated. The pressing pressures were in the range from 86 MPa to 150 MPa, and the sintering process was carried out at 1190 °C for 40 minutes of passing time, which corresponds to the two hours of sintering at given temperatures. The sintered samples have been exposed to a quantitative microstructural analysis in reference to the grain sizes and porosity characteristics. The section area (A), perimeter (L_P), form factor (f), numerical density (N_A) and volume fraction of pores (V_V) have been determined. This analysis has shown that a slight tendency of equalizing of grain sizes is associated with the increase of initial pressing pressure. SEM microscopy has been used also for detailed examination of microstructure. The relative capacitance and resistance in function on pressure have been examined also.

Key words: barium titanate, electrical properties, grain size, mathematical modelling, microstructure, sintering.

INTRODUCTION

Recently there has been of great importance the investigation of complex and comprehensive behaviour of barium titanate ceramics, especially from the point of view of their electrical characteristics (Semiconducting Barium Titanate, 1977; Daniels et al., 1978/79; Mader et al., 1987). Detailed investigation of the relations between microstructure which is defined through the sintering process, as well as by appropriate additives, may provide better understanding of their electrical properties. A great majority of papers concern the investigation of barium titanate with additives which enhance either sintering rate, or densification. Some of them are used to adjust the Curie point and others to promote the semiconducting properties (Wang and Umeya, 1990). In this paper a high purity commercial powder of $BaTiO_3$ was used to examine the relation between technological parameters of processing with microstructure and related electrical properties. Regarding that the pressing process could be treated as a initial process of an overall sintering procedure, the effect of the consolidation of the starting powders has great importance bearing in mind that the deformations and stresses could be introduced into the grains. The grain sizes and consequently the grain boundary area have significant influence on the resistivity of given material. The porosity, which is dependent on the initial pressing pressure, as well as on the sintering process, is also one of the main factors which contribute to the complexity of $BaTiO_3$ behaviour. The processing and development of $BaTiO_3$ components could be improved by using mathematical modelling which requires a definition of a dynamic system with recurrent coupling (Jordović et al., 1993) what lead down to the optimization of $BaTiO_3$ structure.

EXPERIMENTAL

The specimens for this study were prepared from Murata barium titanate powder of the following composition: 65.24% BaO and 34.70% TiO_2. The powder consists of agglomerates range from 10 μm to 120 μm. In order to investigate the influence of the green density on the electrical characteristics and grain growth the following pressures have been used: 86, 105, 130 and 150 MPa. The specimens were sintered in a tunnel furnace type "CT-10 MURATA" at 1190 °C for 120 minutes. Electrical properties, the capacitance and electrical resistance, were measured on the specially prepared components, based on sintered samples, using "Hewlett Packard" device (Saymaprasad et al., 1987; DeHoff, 1987; Mitić, 1989).

In this experimental study the microstructural investigation by optical and SEM microscopy have been done, with a special attention to the metallographic analysis. The measurements were carried out on polished and etched surfaces using a semiautomatic device for quantitative analysis "MOP - Videoplan - Kontron" with automatic data processing.

This analysis provides the investigation of a great number of samples and view fields so that reliable statistical data have been obtained. In Tbl. 1. the minimal, maximal and average grain and pore sizes are given.

Table 1. The grain size as a function of pressure

Pressing pressure (MPa)	Grain size l(μm)		
	l_{min}	l_{max}	l
86	0.36	4.1	0.89
105	0.17	3.5	0.87
130	0.17	2.98	0.62
150	0.21	2.8	0.61

RESULTS AND DISCUSSION

For the analysis of grain size distribution as a function of pressing pressure the measuring method of intercept has been used. Regarding that the grain sizes ranged in the intervals of 0-3 μm, the optimal number of classes was selected with the 0.2 μm of width. For such selected classes, the analysis of absolute frequencies of appearance inside each of individual class has been done. Log normal distribution has been assumed for description of frequencies distributions and could be done in following form:

$$g(l)=\frac{1}{\sigma\cdot l\sqrt{2\pi}}\cdot e^{-\frac{(\ln l-\ln\bar{l})^2}{2\cdot\sigma^2}} \qquad (1)$$

where σ represent a measure of dispersion and l is a grain size diameter.

The values of σ and l are given in Tbl. 2. In Fig. 1 the density function dependence on initial pressing pressure have been shown. As could be seen from the given results, for approximated values of dispersion σ, the average grain size diameter is a direct function of pressing pressure. If the pressing pressure increases the average grain size become smaller, that is manifested by moving the maximum of the curves to the left, towards lesser grain size diameters, as could be seen also from Tbl. 2.

Table 2. The calculated values for grain size $\bar{l}$ and dispersion values σ in function on pressing pressure

Pressing pressure (MPa)	$\bar{l}$ (μm)	σ
86	0.73	0.46
105	0.65	0.43
130	0.54	0.41
150	0.50	0.39

It is noticeable that with the increasing of pressing pressure the average grain size decreases and the frequency of small grains become greater, at the same time the porosity was insignificantly changed.

It is obvious that the average grain size and related distribution are more similar for 86 MPa and 105 MPa, compared to that of 130 MPa and 150 MPa. Namely, with the increasing of pressure, the classes with smaller grain sizes were more represented on account of the decrease of classes with greater grain sizes. However, bearing in mind the differences between the minimal and maximal grain sizes, it could be noticed, that generally, the grain sizes are very equalized, although grain size distribution equalizing has a very weak tendency.

Relationships between the grain sizes diameter for classes of maximal frequencies and pressing pressures, indicate that the grain size is intensively changing up to the pressure of 105 MPa and after that remains constant. Therefore, this value of pressure for given sintering conditions, represents a critical pressing pressure in reference to the grain growth for pure barium titanate.

In this study the dependence of capacitance and resistance on pressing pressure has been investigated also. As could be seen in Fig. 2 the capacitance does not change significantly in the temperature range from 20 °C to 85 °C, indicating the temperature - stable dielectric behaviour of barium titanate. Regarding the dependence of electrical resistance on pressing pressure (Fig. 3) the effect similar to that one of PTCR effects could be recognized. For relatively high pressing pressure of 150 MPa (Fig. 3a) this phenomenon is diminished and NTCR effect is prevailing.

From the results obtained, it is evident that depending on the pressure and sintered conditions, the corresponding resistance can be

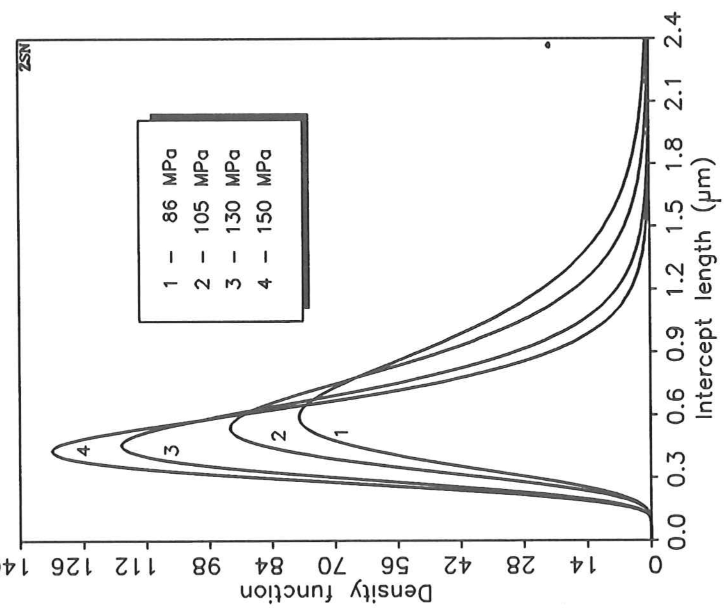

Fig.1.Calculated grain size distributions

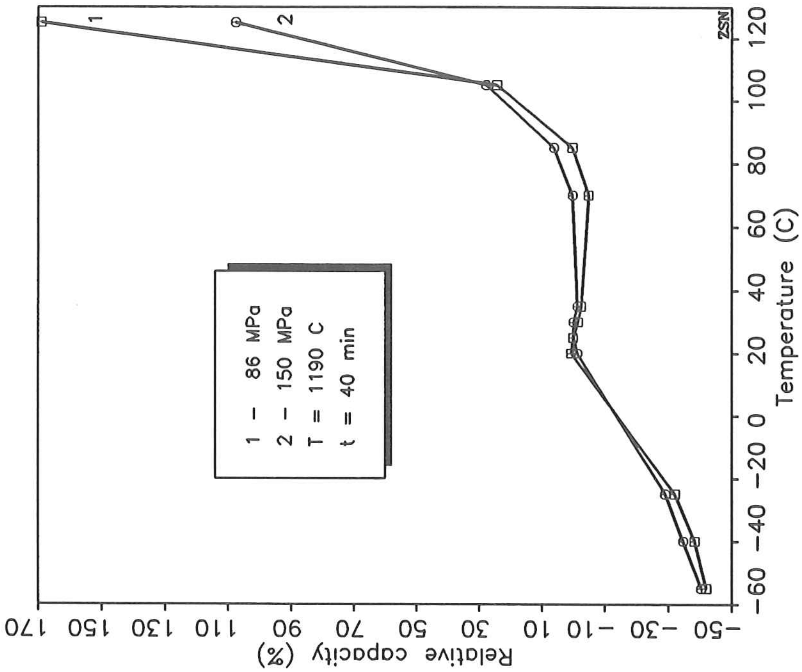

Fig.2.Measured relative capacitance as function of temperature and pressing pressure

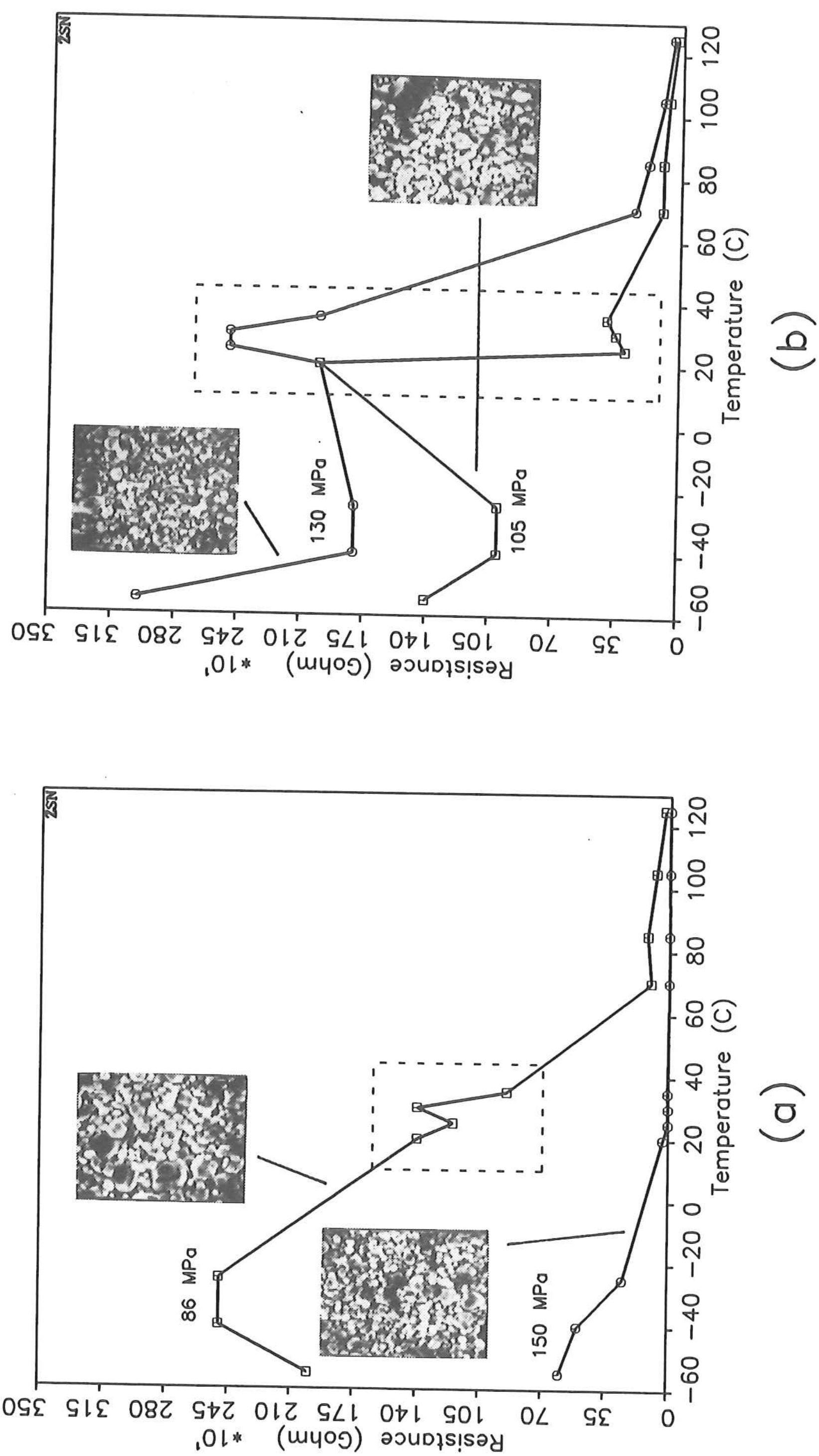

Fig.3. Measured resistance as function of temperature and pressing pressure with SEM microstructures of $BaTiO_3$ - ceramic samples (x5000)

achieved. By increasing the contacts area of grains and due to the uniformity of grain sizes the resistance of the specimens is decreased.

CONCLUSION

In samples sintered at 1190 °C for two hours the normal grain growth is observed independent from initial pressing pressure, although the average grain size decreases as the pressure is rising. For a given sintering conditions, a critical pressing pressure of 105 MPa pointed out that above this pressure the grain growth is negligible. At lower pressures, the results, obtained from electrical resistance dependence on pressures, show the effects with is very similar to that one for PTCR effect. This similarity is diminished at higher pressures. Capacitance do not change significantly in the wide region of temperatures, indicating the temperature - stable dielectric behaviour of barium titanate.

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