Analogy between Mechanical and Dielectric Strength Distributions in BaTiO₃ Thick Films Prepared under the Different Processing Conditions

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調製条件の異なるチタン酸バリウム厚膜における機械強度・絶縁強度分布の相関

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Mechanical and dielectric strength were measured for three types of BaTiO₃ thick films prepared under the different processing conditions. In any case, Weibull moduli for both strength distributions were identical in each type of films, though they differed among three types. The results indicated that dielectric strength measurement is a promising alternative method to distinguish the structural reliability change as influenced by processing parameters before firing within the framework of the same fabrication method.

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1. Introduction

Since Yamashita et al.¹⁾ showed an analogy between mechanical and dielectric strength distributions in BaTiO₃ ceramics, various works have been carried out in an attempt to use the dielectric breakdown test as an alternative method to evaluate mechanical strength reliability.²⁾⁻⁴⁾ The present authors have already reported that the analogy between the two strength distributions should be stemmed from the similar role of microstructure for both failures.²⁾

Yamashita et al.¹⁾ fabricated BaTiO₃ ceramics by hot-pressing and used various kinds of additives to control the microstructure, while we fabricated BaTiO₃ thick films with no additives by a doctor blade method and altered only the sintering temperature to vary the microstructure.²⁾ The fact to be noticed here is that the Weibull moduli (m) of both mechanical and dielectric strengths were ~15 for hot-pressed samples¹⁾ and ~4 for thick films²⁾ in spite of the microstructure variation. Thus dielectric strength measuring method seems to be a promising alternative

method to distinguish the structural reliability of BaTiO₃ ceramics obtained from different methods of fabrication. But it remains to be examined if this method could be applied to the case where the Weibull modulus of mechanical strengths varies according to the changes in processing parameters before firing within the framework of the same fabrication method. In this study BaTiO₃ thick films fabricated from green sheets with different homogeneity were employed to examine the applicability of this alternative reliability estimating method.

2. Experimental procedure

Modified doctor blade method previously reported⁵⁾ was adopted in order to fabricate BaTiO₃ green sheets. Homogeneity of the green sheet was controlled by changing the mixing time of powder and organic binder (methyl cellulose aqueous solution), whose mixing ratio was fixed. One slurry was mixed for 15 min and another for 48 h, while sintering temperature and time were fixed as 1350°C and 3 h respectively. To make comparison, a commercial BaTiO₃ green sheet containing dispersing agent was also sintered at 1350°C for 3 h.

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The obtained BaTiO₃ thick films (A : mixed for 15 min, B : mixed for 48 h, C : commercial) were cut into rectangular specimens of 5 by 20 by 0.2 mm for mechanical and dielectric strength tests. A three-point flexure method was used to measure the bending strength and D. C. dielectric breakdown measurement was carried out with the same samples.²⁾ All the measurements were made at room temperature.

To compare both mechanical and dielectric strength distributions on the same scale, the following equations proposed by Yamashita et al.¹⁾ were used.

$$P_1 = 1 - \exp\{-(E_b/E)^{m_1}(E/E_0)^{m_1}V_1\} \quad (1)$$

$$P_2 = 1 - \exp\{-(\sigma_f/\sigma)^{m_2}(\sigma/\sigma_0)^{m_2}V_2\} \quad (2)$$

where m_1 , m_2 are Weibull parameters, E , σ average strengths, E_0 , σ_0 scale parameters and V_1 , V_2 effective volumes for the respective strength, respectively. These equations are based on the familiar two parameter Weibull distribution. Cumulative failure probabilities, P_1 and P_2 , were calculated using the mean rank method.⁶⁾

Scanning electron microscope (SEM) observations were performed to determine the grain size distributions for each specimen.

3. Results and discussion

Figures 1, 2 and 3 show the set of data for mechanical (○) and dielectric (●) strengths obtained for each specimen group. Both mechanical strength and Weibull modulus increase in the order of the specimens A, B and C. Rogeaux et al.⁷⁾ showed that ultrasonic (US) assistance to powder compaction is useful for increasing the

Weibull modulus and the average mechanical strength of sintered alumina. US assistance enhanced the rearrangement of the powder particles, which resulted in eliminating the badly compacted zone. The results obtained in the present study might be interpreted in the same way : long time mixing and dispersing agent contained in sample C would have given rise to closer compaction of BaTiO₃ powder particles in the green sheets.

It is clearly seen in the figures that the distribution shapes of both mechanical and dielectric strengths are very close to each other. Weibull moduli of mechanical and dielectric

Fig. 2. Weibull plots of mechanical and dielectric strengths for the specimens B ; 48 h mixing.
○ : mechanical strength, ● : dielectric strength

Fig. 1. Weibull plots of mechanical and dielectric strengths for the specimens A ; 15 min mixing.
○ : mechanical strength, ● : dielectric strength

Fig. 3. Weibull plots of mechanical and dielectric strengths for the specimens C ; from commercial green sheets.
○ : mechanical strength, ● : dielectric strength

Table 1. Average mechanical and dielectric strengths, and their Weibull modulus for BaTiO₃ thick films.

Specimen type	A	B	C
Mechanical strength (kgf/mm ²)	3.4	5.3	6.1
Weibull modulus	2.4	4.8	7.9
Dielectric strength (kV/cm)	63	81	93
Weibull modulus	2.6	4.3	7.9

specimen type	A	B	C
average grain size $\bar{D}$ (μm)	3.1	1.9	1.4
standard deviation S	0.37	0.31	0.25
coefficient of variation $C = S/\ln\bar{D}$	0.32	0.48	0.14

Fig. 4. Logarithmic grain size distributions and other statistical data for BaTiO₃ thick films: (a) specimens A (15 min mixing), (b) specimen B (48 h mixing), and (c) specimens C (commercial).

strengths were calculated by a least squares method and are summarized in **Table 1**. In any case, Weibull moduli for both strength distributions appear to be almost identical in each group, though they differ among three types of specimens derived from different mixing conditions. These results suggest that the dielectric strength measuring method could detect the structural reliability change accompanied by the change in processing parameters within the framework of the same fabrication method.

From SEM observations, logarithmic grain size distribution was calculated for each specimen group (A, B, C). Average grain size, stan-

dard deviation and coefficient of variation are summarized in **Fig. 4**.

Both mechanical and dielectric strengths increase with a decrease in average grain size. This grain size dependence in mechanical strength can be explained by means of the well known Petch equation.⁸⁾ Inverse grain size dependence of dielectric strength of MgO has also been reported.⁹⁾

Comparison of the data given in **Table 1** and **Fig. 4** would suggest at first sight that the Weibull modulus increases with both decreasing average grain size and standard deviation of grain size distribution. However the coefficient of variation

is statistically a more fundamental parameter to describe the grain size distribution, and this should be compared with the Weibull modulus. It is noted that there is no positive correlation between them. Hence, further work will be necessary to clarify the most decisive factor to give the coincidence in both strength distributions.

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