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Strength distribution of titania ceramics after high-voltage screening

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Screening fields, at or below which 10%, 30% and 50% of TiO_2 ceramic samples would break electrically, were applied to samples which show single-mode strength distribution. After these high-voltage screenings, the surviving samples were subjected to mechanical strength measurement and the resultant strength distributions were compared with the original distributions. This high-voltage screening was also applied to model ceramics samples composed of titania with two different relative densities. The effect of high-voltage screening and the correlation between mechanical and dielectric strengths are discussed.

1. Introduction

The analogy between mechanical and dielectric strength distributions for several materials has been reported previously [1–4]. On the basis of these results, an alternative method has been proposed to estimate the structural reliability of a dielectric ceramic. Beauchamp [5] and Suzuki *et al.* [6], also reported analogies of the microstructure dependence of mechanical and dielectric failures. Such an analogy in both strength distributions is derived from the coincidence of weak spot distributions for both failures, indicating the coincidence of the starting points of both failures when fracture stress and breakdown field have been applied to the same area.

If the microstructure played a similar role for both failures, not only the strength distribution but also the strengths themselves would show a positive correlation. In other words, a ceramic part with high dielectric strength also shows high mechanical strength and a part with low dielectric strength would show low mechanical strength. Then the probability of high-voltage screening on dielectric ceramics can be expected. In the conventional screening method, final products with a low mechanical strength can be eliminated by applying stress, taking into account the actual stress in service. This high-voltage screening might be advantageous because it only requires the attachment of electrodes without processing a rectangular shape.

In the present study, screening fields at or below which 10%, 30%, 50% of TiO_2 ceramic samples would break electrically, were applied to the samples which show single-mode strength distribution. After the high-voltage screenings, the surviving samples were subjected to mechanical strength measurement and the resultant strength distributions were compared to the original distributions. The effect of high-voltage screening and the correlation between mechanical and dielectric strengths are discussed. This high-voltage screening was also applied to model ceramic samples composed of titania with two different relative densities.

2. Experimental procedure

2.1. Sample preparation

Titanium dioxide ceramics were employed because they could be easily broken electrically at room temperature. Titanium dioxide powder (Fuji Titanium Co. Ltd, Tokyo, Japan, TP-34D206, purity 99.99%, particle size 0.5 μm) was used as starting material [7]. Powder compacts were first fabricated by uniaxial pressing in a rectangular die ($15 \times 5 \times 35$ mm^3, inner dimensions) at 95 MPa for 60 s. After hydrostatic pressing under 100 MPa for 3 min, the resultant compact bodies were sintered at 1450 °C for 3 h in air. The sintered bodies thus obtained had a relative density of about 90.3% (denoted the 90% sample). Low-density samples were fabricated by changing the sintering temperature to 1100 °C, producing a ceramic with a relative density of 58.4% (denoted the 60% sample). In accordance with our SEM observation, average grain sizes were approximately 1 and 5 μm for 1100 and 1450 °C sintered samples, respectively.

Test pieces commonly used for both strength measurements and screening tests were prepared from bulk ceramics as follows. First, the surface layers of bulk ceramics were removed with no. 400 SiC abrasive paper (Nippon Coated Abrasive Co. Ltd, Aichi-ken, Japan, C947). The ground samples were cut into rectangular bars of $12 \times 4.0 \times 0.3$ mm^3 with a precision cutting machine (Maruto Co. Ltd, Tokyo, Japan, MC-603).

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2.2. Strength measurement

Silver electrodes with a diameter of 3.5 mm were evaporated approximately at the centre of both sides of all test bars. These electrodes were made to have diffused edges to prevent edge-field concentration.

Mechanical strength was measured by three-point bending (10 mm span) with a crosshead speed of 0.2 mm min^{-1} on samples with electrodes. Taking into account the strength compared to an electrically screened sample, the maximum bending stress was applied at the centre-line of the electrode. The fractured samples were also subject to dielectric breakdown measurement for the purpose of a distribution comparison. Silver electrodes were similarly evaporated and the d.c. voltage was increased on the specimen placed in silicone oil to prevent surface discharge. The breakdown field at which a current abruptly increased was regarded as the dielectric strength, E_b.

Distribution of mechanical strength before and after screening, as well as the dielectric strength, were estimated using the two-parameter Weibull distribution function [8], as follows

$$P = 1 - \exp[-(s/s_0)^m V] \quad (1)$$

where s is the mechanical or dielectric strength, s_0, m, V are the scale parameter, shape parameter (Weibull modulus), and effective volume for each strength, respectively. Cumulative failure probability, P, was calculated using the mean rank method.

2.3. High-voltage screening on samples with single-mode strength distribution

Three sets of 90% titania ceramics were used to investigate high-voltage screening on samples with single-mode strength distribution. This experimental procedure is schematically illustrated in Fig. 1.

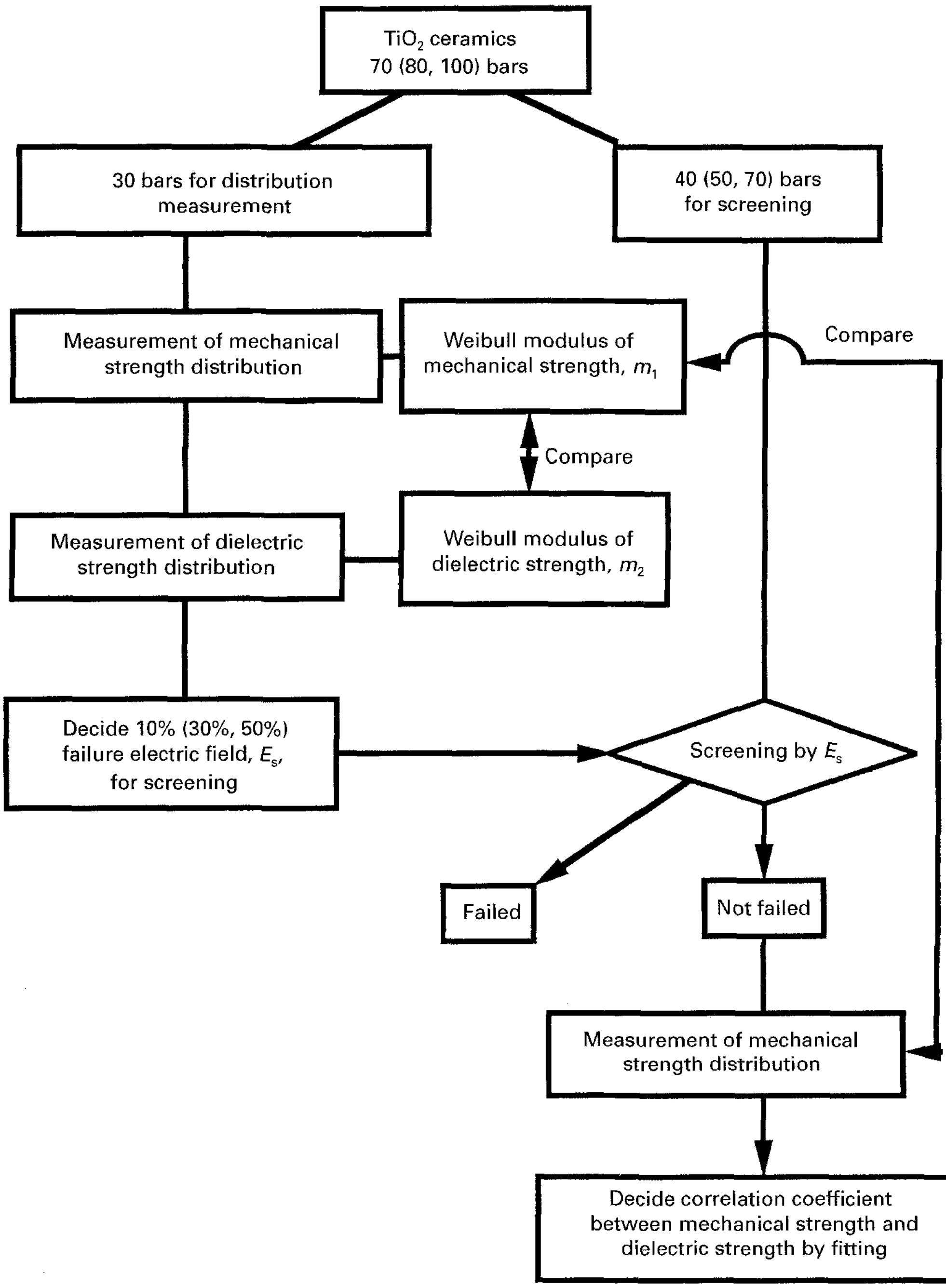

Figure 1 Schematic diagram of the high-voltage screening experiment on a sample with a single-mode strength distribution.

Three groups composing 70, 80 and 100 test bars, were named Groups A, B, and C, respectively. From each group 30 bars were picked for comparison of both strength distributions. The residual 40, 50 and 70 test bars were used for screening. To compare both mechanical and dielectric strength distributions on the same scale, the following equations, proposed by the present authors [1–4] were used

$$P_1 = 1 - \exp[-(E_b/E_{av})^{m_1}(E_{av}/E_0)^{m_1}V_1] \quad (2)$$

$$P_2 = 1 - \exp[-(\sigma_f/\sigma_{av})^{m_2}(\sigma_{av}/\sigma_0)^{m_2}V_2] \quad (3)$$

where m_1, m_2 are Weibull moduli, E_{av}, σ_{av} are average strengths, E_0, S_0 are scale parameters and V_1, V_2 are effective volumes for the dielectric and mechanical strengths, respectively. These equations are based on the familiar two-parameter Weibull distribution [8]. Cumulative failure probabilities, P_1 and P_2, were calculated using the mean rank method.

From the dielectric strength distributions of groups A, B, and C, the electric fields at or below which 10%, 30%, and 50% of samples would break electrically were calculated, and these values were taken to be each screening field, E_S. Similar to the dielectric breakdown test, the electric field was increased up to E_S and samples which broke during the screening test were eliminated. Mechanical strength was measured on samples which survived.

2.4. High-voltage screening on samples with biomodal strength distribution

High-voltage screening was investigated for samples with a bimodal strength distribution. The experimental procedure is schematically illustrated in Fig. 2. Two sample groups, composing 20 pieces of 60% test bars and 20 pieces of 90% test bars, were prepared and named groups C and D, respectively. Group C was used for strength measurement. The other group D, was used to investigate the effect of high-voltage screening.

First, mechanical strengths were measured on group C, where the maximum bending stress was also applied at the centre-line of the electrode. In order to determine the screening field, dielectric breakdown measurement was conducted on fractured samples which had mechanical strengths in the lower strength range. The obtained dielectric strengths were statistically treated to determine the screening field, E_S, at or below which all the 60% relative density samples would break.

Similar to the dielectric breakdown test, electric field was increased up to E_S for group D, and samples broken during this screening test were eliminated. Three-point bending strength was measured on the surviving samples and strength distribution was compared with that of group C.

3. Results and discussion

3.1. Comparison of mechanical and dielectric strength distributions

A comparison of mechanical and dielectric strength distributions of group C is shown in Fig. 3. Both Weibull plots show good linearity (correlation coefficient $r > 0.98$), indicating that the scattering in both data sets can be expressed by a single-mode Weibull distribution function and that both mechanical and dielectric failures obey the weakest-link hypothesis.

It can also be seen that the distribution shapes of both strengths are very close to each other. Weibull moduli of dielectric and mechanical strengths were calculated by a least-squares method to be 10.9 and 11.4, respectively. In this case, Weibull moduli for dielectric and mechanical strength distribution are considered to be substantially the same, as already been reported for various $BaTiO_3$, TiO_2, and MgO ceramics [1–4]. Weibull moduli of dielectric and mechanical strengths are listed in Table I together with the results for other group. Weibull moduli for both strength distributions appear to be almost identical in each group. In addition, the Weibull moduli themselves are almost the same for all groups, indicating the reproducibility of the distributions as well as the ceramic microstructure. The electric fields at or below which 10%, 30%, and 50% samples break in each group are also listed in Table I.

3.2. Effect of high-voltage screening on a sample with single-mode strength distribution

The results of mechanical strength distribution after high-voltage screening with $E_{S_{10}}$ are shown in Fig. 4a, together with original strength distributions. Six out of 40 samples were eliminated by high-voltage screening with $E_{S_{10}}$. After screening, the Weibull plots bend to become a convex curve while plots in the high-strength region remain almost the same. It can be seen that the low-strength region shifts in the high-strength direction, indicating that the low-strength samples were eliminated by the high-voltage screening.

The failure probability after screening, P_a, by stress is usually expressed as [9]

$$P_a = \frac{P_{total} - P_S}{1 - P_S} \quad (4)$$

where P_{total} is the failure probability without screening and P_S is the failure probability with a screening stress. According to this equation, the probability of theoretical failure after screening by a stress $P_S = 0.1$ is drawn in the same figure as a convex thin solid line.

In the case of stress screening, no sample fell below the screening strength unless damage was introduced by the screening stress. In the case of high-voltage screening, however, some samples had a strength lower than the stress at which 10% of the sample should have failed, $\sigma_{S_{10}}$, while almost 10% of the samples were eliminated by the electric field, $E_{S_{10}}$.

The other screening data with $E_{S_{30}}$ and $E_{S_{50}}$ are illustrated in Fig. 4b and c, together with the original lines and the theoretical lines with stress screening, $\sigma_{S_{30}}$, $\sigma_{S_{50}}$. Similar to $E_{S_{10}}$ screening, the Weibull plots bend to become convex curves, indicating that low-strength samples were eliminated by the high-voltage screening. The degree of the shift of low-strength

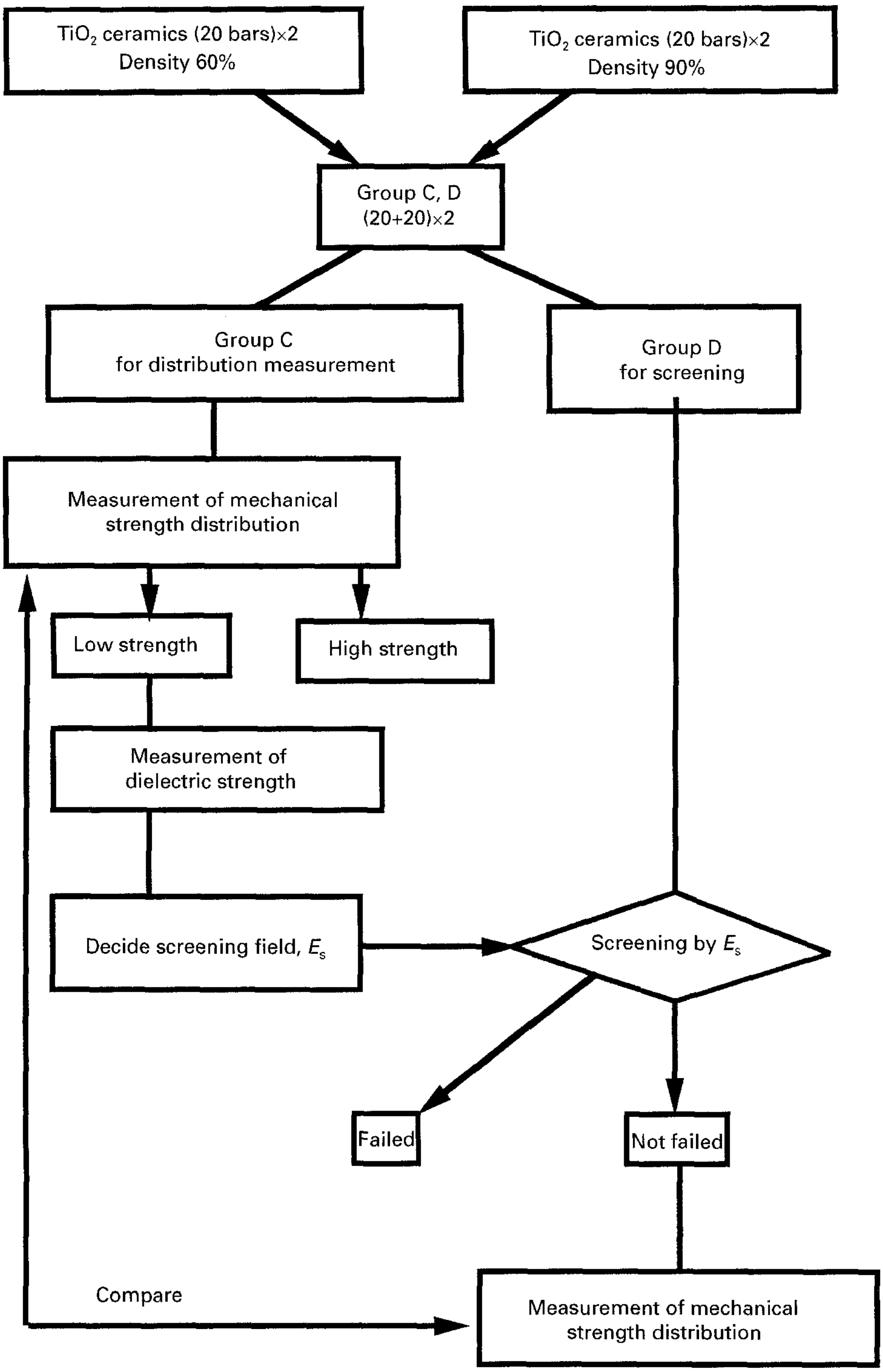

Figure 2 Schematic diagram of the high-voltage screening experiment on a sample with a bimodal strength distribution.

sites is in the order $E_{S_{10}} < E_{S_{30}} < E_{S_{50}}$. As a result, the distribution width decreases with screening field. The theoretical line, however, is higher than the screened data, so the screening data are located between the original line and theoretical line with stress.

3.3. High voltage screening on samples with bimodal strength distribution

Weibull plots of mechanical strength in group C are illustrated in Fig. 5. According to this figure, the plots can be separated into two regions, a high-strength region and a low-strength region. The high-strength region is thought to correspond to 90% density samples, and the low-strength region to 60% density samples. These two peaks in (bimodal) strength distribution can be expressed by a mixed mode Weibull distribution, which corresponds to the situation where one origin governs the fracture of a sample while the fracture origin may differ between samples (depending on their densities, 60% or 90%, in the present case) [10]. In the present case, however, it was difficult to identify the fracture origin, especially in low-density samples. Hence, it remains unknown whether or not only one origin governs the fracture.

The dielectric strength on samples in which mechanical strengths belong to the low-strength region, were measured, and the distribution was estimated. Because the plots showed good linearity, indicating only one origin governs the dielectric breakdown,

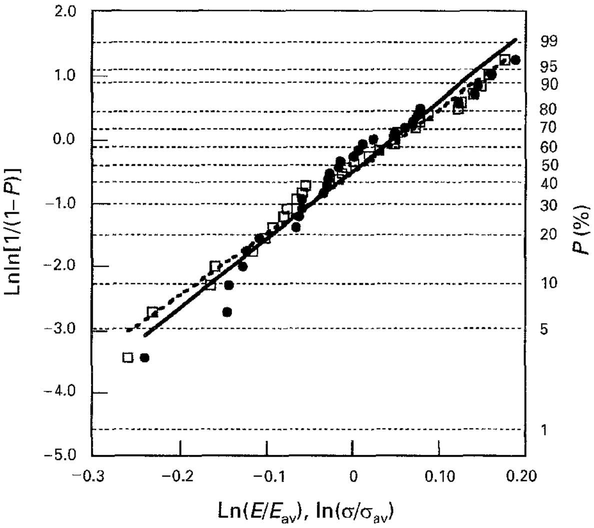

Figure 3 Weibull plots of (●) mechanical and (□) dielectric strengths for TiO_2 ceramics.

TABLE I Weibull moduli of mechanical and dielectric strengths for each group and determined from the screening field

	Group		
	A	B	C
m_1	10.8	10.4	10.9
m_2	9.8	11.3	11.4
E_S (kV cm^{-1})	115	132	198

while change in microstructure before and after dielectric breakdown was not identified. The average dielectric strength, 95% sample breakdown field, and 99% breakdown field are 85, 110 and 125 kV cm^{-1}, respectively. Then the screening field, E_S, at which almost all low mechanical strength samples break, 125 kV cm^{-1}, was used in the following screening test.

Of 40 samples, 22 broke by high-voltage screening up to 125 kV cm^{-1}, indicating that samples belonging to the low-mechanical strength region can be eliminated. The surviving samples were subjected to the three-point fracture test and their mechanical strengths were also treated by Weibull statistics as shown in Fig. 5. The plots of mechanical strength after high-voltage screening show good linearity, indicating they are composed of only high mechanical strength samples. According to the SEM observation, the surviving samples after screening were thought to be 90% density ones.

The slope of the plots of the high-strength region in group C, and that after high-voltage screening, are approximately 1:2. In other words, the plot of mechanical strength after high-voltage screening is equivalent to that of mechanical strength before screening in the high-strength region.

4. Discussion

4.1. Correlation between mechanical and dielectric strength

The reason for the discrepancy between stress screening and high-voltage screening is discussed in relation

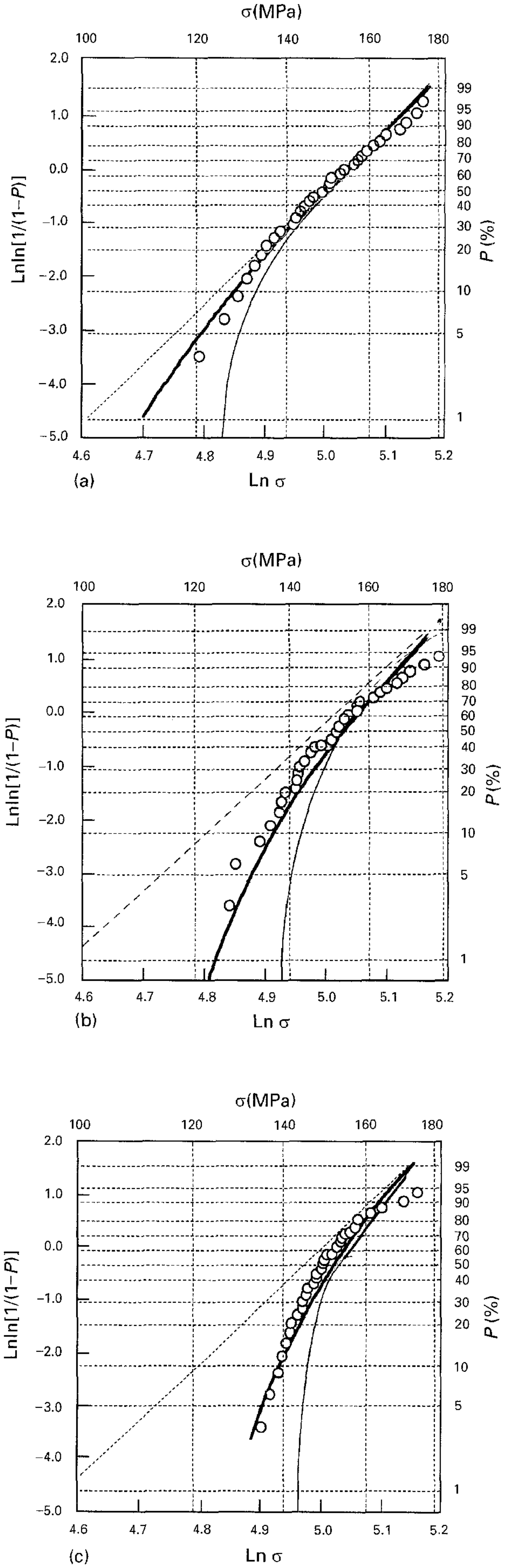

Figure 4 Weibull plots of mechanical strengths for TiO_2 ceramics with single-mode strength distribution (— upper) before and (○) after high-voltage screening with (a) $E_{s_{10}}$, (b) $E_{s_{30}}$ and (c) $E_{s_{50}}$, together with (— lower) the theoretical screening results. (- - -) Original strength distributions.

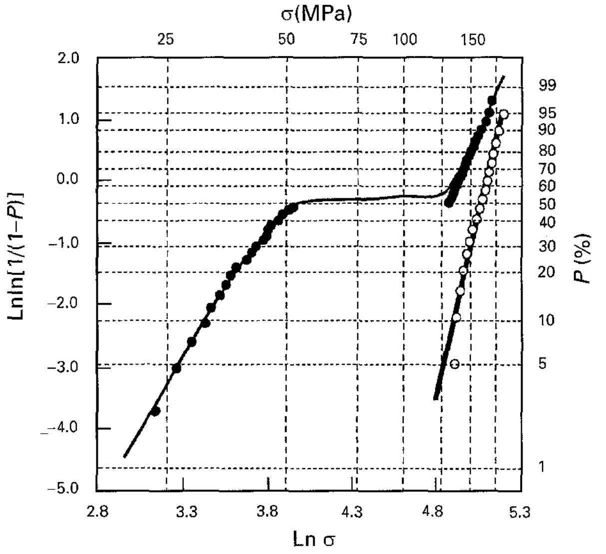

Figure 5 Weibull plots of dielectric strength for TiO_2 ceramics with a bimodal strength distribution (●) before and (○) after high-voltage screening. Correlation between mechanical and dielectric strengths was by computer simulation.

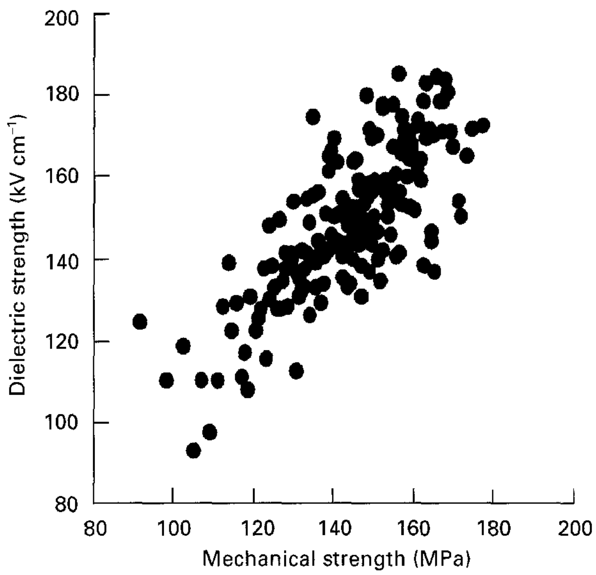

Figure 6 A correlation between mechanical and dielectric strengths with a correlation coefficient of 0.77 and simulated screening results.

to the correlation between mechanical and dielectric strengths.

If there is a perfect correlation between mechanical and dielectric strengths (correlation coefficient = 1), the samples with mechanical strength less than $\sigma_{s_{30}}$ should be eliminated through the high voltage screening with $E_{s_{30}}$. On the other hand, if the correlation is not so perfect (correlation coefficient $\neq$ 1), some mechanically weak samples lower than $\sigma_{s_{30}}$ could exist after high-voltage screening with $E_{s_{30}}$.

There ought to be a positive correlation between mechanical and dielectric strength with a correlation coefficient less than unity. This would lead to a discrepancy between stress and high-voltage screening while a considerable effect of high-voltage screening is demonstrated. High-voltage screening was then simulated on hypothetical mechanical and dielectric strength pairs, σ_{fi}, E_{bi}, with various correlation coefficients, r.

In total, 100 mechanical strengths were generated to obey the experimental strength distribution. For each mechanical strength, a dielectric strength was allocated to make strength pairs. In this process a certain correlation between mechanical and dielectric strengths was assumed and the set of dielectric strengths were made to obey the experimental dielectric strength distribution. From the 100 strength pairs, pairs with $E_b < E_{s_i}$ (i = 10, 30, and 50) were eliminated and the mechanical strength distribution of the remaining pairs was calculated. The simulated strength distributions with various r were compared to the experimental data. As a result, the simulated line fits best with experiment when $r = 0.77$ in all cases. The resultant calculated lines are drawn in Fig. 4a, b, c and the correlation between mechanical and dielectric strengths with $r = 0.77$ is illustrated in Fig. 6.

The reason why the correlation coefficient between mechanical and dielectric strengths is less than unity is thought to be as follows. The defects which govern both failures are not the same. The effect of a defect on both failures is not equivalent, even if the same defect controls both failures.

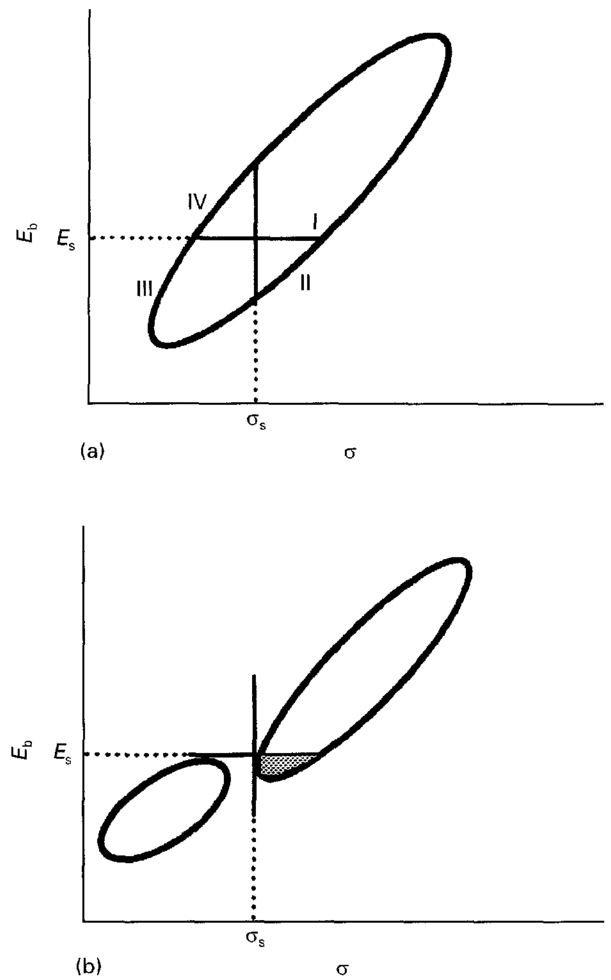

Figure 7 Schematic illustrations of the correlation between mechanical and dielectric strengths. (a) Single-mode strength distribution, (b) bimodal strength distribution.

4.2. Effect of high-voltage screening on samples with bimodal strength distribution

The reason why samples surviving high-voltage screening of model samples with bimodal strength distribution, belong to the high-strength region, is now discussed.

As noted in the previous section, there was a correlation between mechanical and dielectric strengths with correlation coefficient of 0.77. This relation is schematically illustrated in Fig. 7a as a contour line of probability density. In such a case, mechanically weak samples were preferentially eliminated when samples broken by E_S (corresponding to regions II and III) were eliminated. However, mechanically weak samples could survive after screening while their dielectric strengths are large (corresponding to region IV).

In the case of bimodal mechanical strength, however, a contour line of probability density is thought to be composed of two ovals corresponding to high mechanical strength (right) and low mechanical strength (left) as shown in Fig. 7b. Because the low-density sample is thought to have low dielectric strength, the oval for high mechanical strength is located above that for low mechanical strength in the figure. In this case, if E_S is set between the two ovals, the probability density corresponding to region IV in Fig. 7a is very small. As a result, no mechanically weak sample is left after high-voltage screening. However, the small amount of dielectric strength for the high mechanical strength group is smaller than that for the low mechanical strength region (corresponding to the shaded area in Fig. 7b). As a result, only a small number of high mechanical strength samples (two in the present case) could be eliminated by the high-voltage screening.

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