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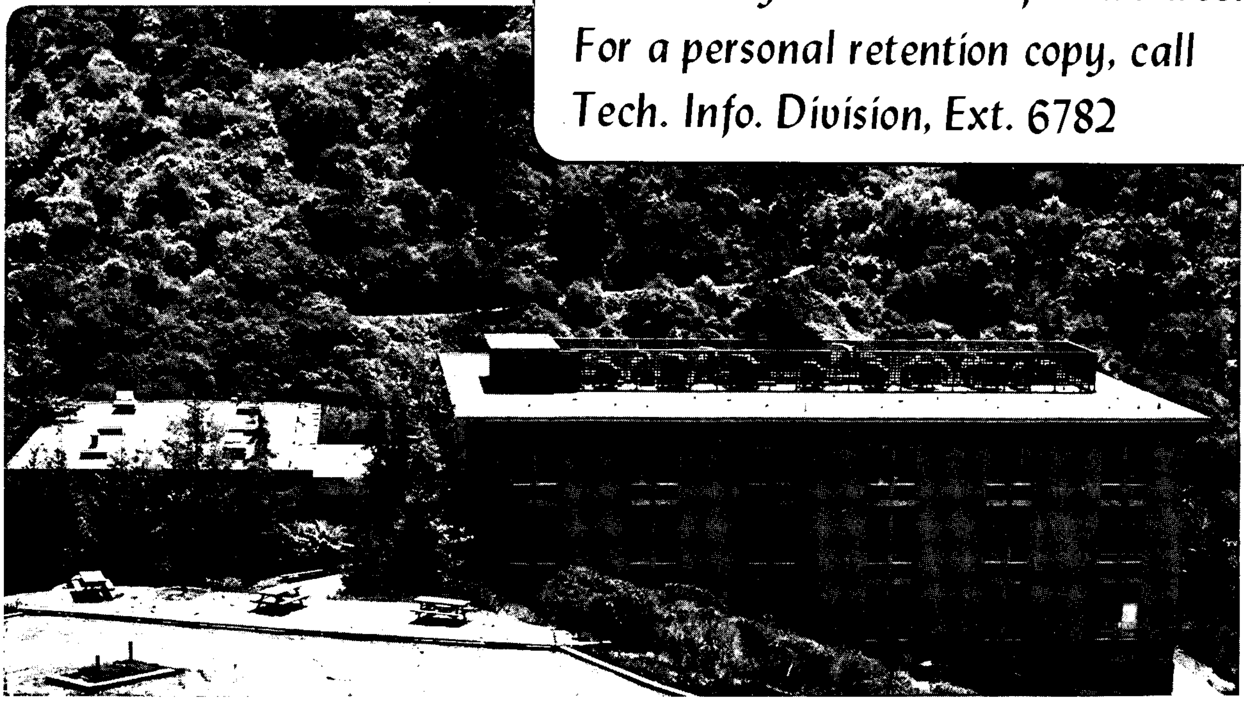

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DISCLAIMER

INFLUENCE OF MICROCRACKING AND SLOW CRACK GROWTH ON PLANAR COUPLING COEFFICIENT IN PZT

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ABSTRACT

The difference in planar coupling coefficient k_p of two PZT specimens with different grain sizes under the same poling treatment was attributed to a greater tendency for microcracking in the large grain size specimens. In the absence of microcracking, as shown in small grain size specimens, the decrease in k_p and failure of the poled specimens is attributed to slow crack growth. The optimum poling conditions for small grain size specimens were evaluated in terms of reducing the slow crack growth effect.

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*Deceased

The planar coupling coefficient, k_p, has been used extensively as a measure of the piezoelectric response of PZT ceramics. It was found that k_p depends on the material parameters[1-3], such as grain size, porosity, chemical composition. Okazaki[3], reported that k_p decreases with increasing porosity and with decreasing grain size, and a space charge model[3,4] has been proposed which is dependent on these characteristics. It was found that for a given specimen k_p depends on the poling field, temperature and time[5-7]. These dependencies varied strongly from one composition to another and were attributed to the nature of the doping species[8] or the Zr/Ti ratio. However, the dependence of k_p on poling conditions was also found to vary considerably for a number of specimens with the same composition but different microstructures.

Takahashi et al.[9] have reported that when undoped $Pb(Zr_{0.52}Ti_{0.48})O_3$ with density 7.7 - 7.8 g/cm^3 and grain size of 22 μm was poled at 100°C with a field of 4 KV/mm, k_p increased as time increased, reached a maximum value of 0.61 when poled for 10 minutes and then decreased to 0.58 when poled for 60 minutes. On poling with a field of 5 KV/mm, k_p increased as time increased and reached a maximum value of 0.56 when poled for 60 minutes. Thus, on increasing the field from 4 KV/mm to 5 KV/mm, k_p dropped for each poling time within the experimental range. The explanation presented by these authors[9] was that the bonding strength between grains is weak for pure materials. They compared the fracture surface of 0.3 wt% Cr doped and undoped specimens and concluded that the bonding strength was indeed weaker for the undoped specimen due to its intergranular fracture compared to the transgranular fracture for the doped specimen.

Webster et al.[10] reported that for PZT with the same composition, small grain size of 2 μm, and nearly the same density of 7.9 g/cm^3, k_p was found to be 0.61 under nearly the same poling condition (100°C, 3.6 KV/mm for 5 minutes). If the space charge model is employed to explain this difference, we would expect that Takahashi's specimen[9], with grain size 10 times larger, would show a higher coupling value. It has been found[11] that k_p can be as high as 0.65 if an improved processing procedure is used. Thus 0.61 is not the limiting value for this material under poling. Purity was not a factor to be considered since both specimens were of similar purity. In order to clarify the grain size effect, we have prepared small grain size PZT specimens with the same composition[9] and studied the dependence of k_p on the poling conditions with an objective of finding the optimum conditions for poling.

EXPERIMENTAL PROCEDURES

99.9% pure reagent grade PbO, ZrO_2 and TiO_2 powders were used to formulate $Pb(Zr_{0.52}Ti_{0.48})O_3$. The mixed powders were calcined at 850°C for 4 hours. Three wt% excess PbO was added to the calcined powder prior to forming and sintering as a liquid phase sintering aid and to counteract the loss of PbO by vaporization during firing. Lead zirconate and 5 wt% zirconia (PZ+Z) powder with particle size less than 60 mesh was used as a packing powder. Sintering was carried out at 1200°C for a period of 8 hours. The detailed fabrication process is described elsewhere.[11] Polished disc specimens with radius 2.0 cm and thickness 0.1 cm were poled under various field, temperature and time conditions and aged for 24 hrs. k_p was then evaluated according to the IRE (1961) standard method.

RESULTS AND DISCUSSION

The results for specimens with a 8.0 g/cm^3 density and 3.5 μm grain size at a poling temperature of 100°C and poling fields of 1.k to 6 KV/mm are shown in Fig. 1. k_p increases with increased poling time and approaches a saturation within a period of 30 minutes. Furthermore, k_p increases monotonically as field increases for a given poling time. The difference in behavior compared with Takahashi's results[9], as pointed out in Table 1, is attributed to the occurrence of microcracking in his large grain size specimens.

Microcracking phonomena in ceramics have been recognized for some time. Intercrystalline stresses result from thermal expansion anisotropy or phase transformation in noncubic polycrystalline materials during cooling and heating and are enhanced with increase of grain size.[12-14] When these stresses are large enough, a tendency for internal microcracking generally occurs in brittle ceramics, increasing with grain size. Kuszyk and Bradt[12] found that microcracking occurred in $MgTi_2O_5$ due to thermal expansion anisotropy only when the grain size was larger than 3 μm. Matsuo and Sasaki[13] demonstrated a similar behavior in lead titanate ceramics; and this effect was observed in barium titanate by Rice et al.[14] If microcracking, or separation at grain boundaries, occurs at larger grain sizes, a space charge is created causing the k_p to decrease.[3] In addition, k_p is known to increase with increase in grain size. Therefore, the actual magnitude of k_p is determined by the competitive effect of grain size itself and the related microcracking.

In the course of poling, the dipole alignment caused by the application of an electric field contributes to the development of internal stress. In the absence of microcracking, as for small grain size specimens, the dipole alignment causes an increase in k_p. Saturation of k_p under a fixed field is expected as the poling time is increased since the maximum degree of dipole alignment is restricted by the field applied (Fig. 1). Large grain size specimens behave similarly if the internal stress contributed by the poling field is not large enough to make a significant contribution to microcracking. If the poling field is increased, however, the (field enhanced) microcracking effect becomes dominant and k_p drops. These correlations are supported by the data presented in Table 1.

Data for small grain specimens under various poling conditions are shown in Table 2. Under severe poling conditions (high temperatures, high fields or long times), k_p decreases and subsequently the specimen fails (breakdowns or cracks) at a dissipation current of approximately 0.4 - 0.5 mA. This behavior under the indicated experimental conditions is postulated to be due to slow crack growth along grain boundaries. Examination by SEM of a specimen poled at 150°C and 3 KV/mm whose dissipation current reached 0.50 mA in less than 30 seconds indicated many small cavities along grain boundaries emerging on the flat surface while none were visible in the unpoled specimens. Mechanical failure of many ceramic materials has been characterized by the propagation of small pre-existing flaws to a critical size under severe experimental conditions.[15-17] An enhanced slow crack growth rate (crack velocity) for PZT has been reported by Bruce et al.[18] on exposure to elevated poling temperatures.

Thus, as seen in Table 2, k_p for small grain specimens is increased more effectively by increasing the poling field rather than raising the poling temperature which would enhance the slow crack growth effect. However, if the temperature is too low, practically no domain alignment process can be realized and a low k_p results. As an example, the specimen poled at 10°C, even under a field of 7 KV/mm, shows a k_p value of only 0.28 and almost no dissipation current across the specimen (Table 2). The effect of temperature on k_p is further shown in Fig. 2. k_p increases rapidly when the field is increased from 2 to 3 KV/mm at 100°C. Similar behavior was also reported for lead titanate ceramics by Ikhegami et al[19]. The optimum poling temperature in this case then is 100°C since it enables faster and easier domain alignment in the apparent absence of any significant slow crack growth phenomena.

In summary, the interpretations of the poling behavior of the specimens are based on their susceptibility to the formation of microcracks during processing and to slow crack growth phenomena which are generated by the poling conditions. These phenomena appear to be minimized by characterized processing to realize a favorable microstructure, specifically a small grain size in this case, and by poling at the lowest possible temperature.

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Table 1

Density (g/cm^3)	Grain Size (μm)	Field (KV/mm)	k_p
7.7-7.8†	22	3	.46
		4	.61
		5	.54
8.0 ††	3.5	3	.49
		4	.58
		5	.60
		6	.61

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† Specimens prepared by Takahashi et al.[9]

†† Specimens prepared in this study.

Poling conditions: 100°C and 10 mins.

Table 2

Poling Temp. (°C)	Field (KV/mm)	Time (mins)	Breakdown	Dissipation Current (mA)	K_p
150	3	<.5	yes	.43	-
140	4	1	yes	.50	-
125	4	4	yes	.48	-
100	6	5	No	.07	.60
100	6	10	No	.11	.62
10	7	5	No	∿0	.28

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*$PZ_{0.52}T_{0.48}$ specimens with 8.0 g/cm^3 density and 3.5 µm grain size.

Figure Captions

Fig. 1. Variations of k_p with poling time for indicated conditions.

Fig. 2. Variations of k_p with poling field for indicated conditions.

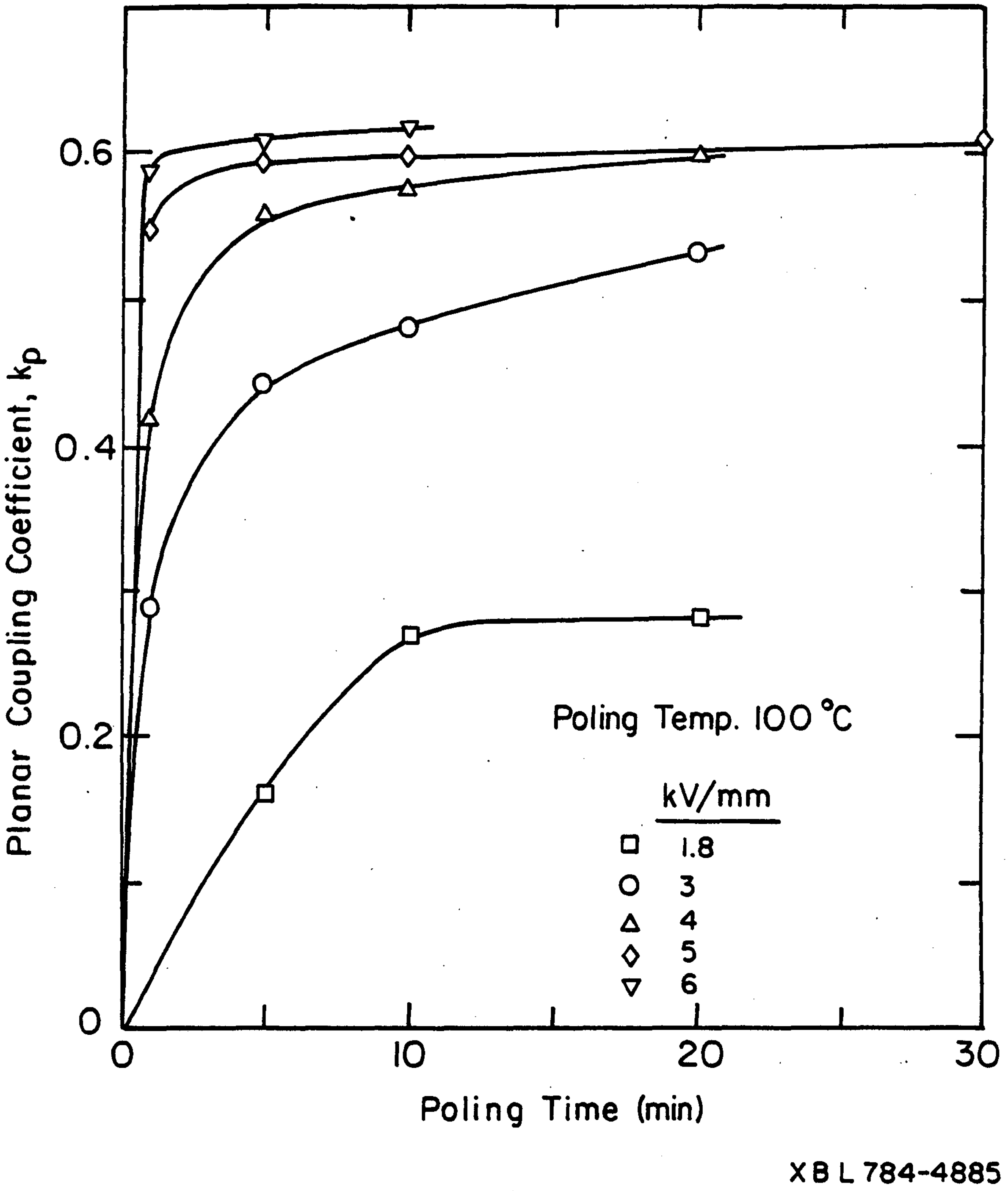
Planar Coupling Coefficient, k_p
0
0.2
0.4
0.6
0
10
20
30
Poling Time (min)
Poling Temp. 100 °C
kV/mm
□ 1.8
○ 3
△ 4
◇ 5
▽ 6
XBL 784-4885

Fig. 1

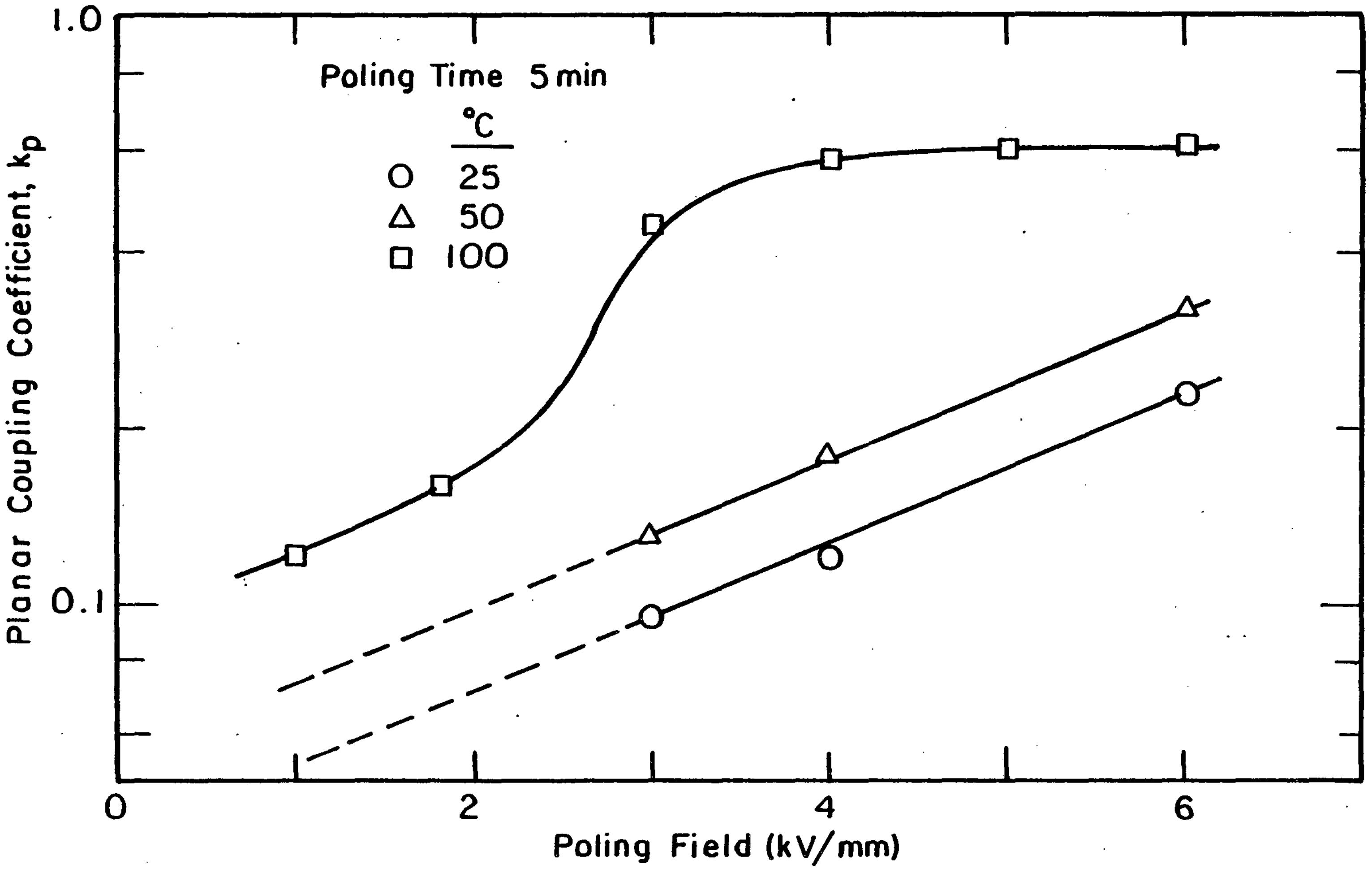

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Fig. 2

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