Crack growth during poling and polarisation reversal in commercial piezoceramics

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The growth of Vicker's cracks during the poling of piezoelectric ceramics was studied for two PZT based compositions with different tetragonal distortion of the perovskite structure. Studies on the two compositions with different poling electric fields and similar initial crack lengths showed that crack growth was not proportional to the induced stress free longitudinal strain by 90° domain reorientation. This observation is not consistent with the previously proposed mechanism of crack growth based on a strain mismatch between the material at the crack flanks and the rest of the ceramic. This mismatch was proposed to occur because of the reduction of the electric field within the crack flanks. The reasons for the disappearance of the mismatch are discussed here, and an alternative mechanism of crack growth consistent with our results is proposed, which takes into account the electrically induced stress gradient at the crack tip, produced by the piezoelectric effect. Cracks were also found to grow during a small number of subsequent polarisation reversals, the explanation of which must be due to some other additional effect.

keywords: Piezoceramics, crack growth

Crecimiento de grieta en piezocerámicas comerciales durante en polarizado

Se ha estudiado el crecimiento de grietas Vicker durante la polarización de cerámicas piezoeléctricas para dos modificaciones del PZT con distinta distorsión tetragonal de la estructura perovskita. Los resultados en función del campo eléctrico de polarización y la longitud inicial de la grieta muestran que el crecimiento no es proporcional a la deformación libre de tensión inducida por orientación de dominios de 90°. Esta observación no es consistente con el mecanismo de crecimiento de grieta propuesto anteriormente, basado en el desajuste de deformación entre el material a los flancos de la grieta y el resto de la cerámica. Este desajuste se producía por la atenuación del campo eléctrico en los flancos de la grieta. Se discute la causa de la desaparición de este desajuste de deformación, y se propone un mecanismo alternativo de crecimiento de grieta consistente con los resultados obtenidos, que tiene en cuenta el gradiente de tensión en la punta producido por el efecto piezoeléctrico. También se ha observado crecimiento de la grieta durante un pequeño número de conmutaciones de la polarización realizadas a continuación del polarizado inicial, fenómeno que debe estar relacionado con algún otro efecto adicional.

Palabras clave: Piezocerámicas, crecimiento de grieta

1. INTRODUCTION

Mechanical issues in piezoelectric and electrostrictive ceramics are becoming increasingly important with the development of high strain actuators (1). Pre-existing processing flaws are liable to interact with the electric fields during the device poling or driving leading to failure (2). Furthermore, additional flaws may appear under the applied field as a result of the large stresses developed (3-5).

Most of the research on the interaction of flaws with electric fields has been focused on surface flaws and fields well above the coercive field of the ceramic, E_C (6-8). Vicker's cracks oriented perpendicular to the electric field have been shown to grow during cyclic electric loading at fields above E_C in lanthanum modified lead zirconate titanate (PLZT) ceramics (6,7) and in lead zirconate titanate (PZT) multilayer actuators (8). Same cracks have been also shown to grow already during the poling of the PZT actuators (8). All growth phenomena were related to a strain mismatch between the cracks flanks and, either contact points along the crack profile (6,7), or the rest of the ceramic (7,8). The mismatch was proposed to build up because

of the reduction of the electric field within the crack flanks to a value below the coercive field. 90° domain reorientation was assumed not to occur at the flanks (untransformed region), but to occur either at the contact points or at the rest of the ceramic (transformed region), leading to the appearance of a tensile stress at the crack faces, which makes the crack grow. If this mechanism is correct, there would be a dependence of the crack growth on the level of strain mismatch, and therefore, on the free stress induced strain. Different free stress strains can be achieved in piezoceramics by using different electric fields. The range may be further widened by using materials with different compositions, and therefore, different crystallographic distortion.

This communication comprises our own experiments on Vicker's crack growth during poling and subsequent polarisation reversals. By using two PZT based compositions, both with the tetragonal structure but different tetragonal distortion, and an increasing poling electric field, crack growth was measured in ceramics in which an increasing free stress longi-

Figure 1. (a) Vicker's indent with 10 Kg for a 4D specimen, and direction of the electric field. One of the former cracks oriented perpendicular to the field (b) before, and (c) after a poling treatment at 3.5 kV mm^{-1} .

tudinal strain was developed. By using different indenter loads, the length of the initial cracks was also varied. The experiments were performed in bars with an appropriate geometry for the thickness excitation of the length resonance. All, the dielectric permittivity of the bars, and the different parameters given by the resonance technique (9), mainly the compliance s_{11} , were monitored during the experiments as an indirect way of determining that the strains were related to 90° domain reorientation, and not to the feasible appearance of microcracks.

2. EXPERIMENTAL

Table I lists some of the properties of the materials, as provided by the manufacturer. The two materials studied were PZT based compositions, PZT4D and PZT5H, representative of the so called "hard", and "soft" piezoceramics, respectively. The crystalline phases presented by the ceramic were investigated by X-ray diffraction. The ceramics were received as poled electroded plates, from which bars $25 \times 5 \text{ mm}^3$ were cut. One of the largest non-electroded sides of each bar was polished with $1 \mu\text{m}$ diamond. The bars were thermally depoled by heating well above the Curie Temperature (see Table I). After the thermal treatment, a set of four Vicker's indents was introduced into the polished surface with a load of 10 Kg. Two additional sets were introduced in a small number of specimens with loads of 2.5 and 5 Kg, respectively. The indents were placed so that the cracks, emanating from the corners of the indentation impression, were parallel and perpendicular to the electrodes, and therefore, to the electric field in the experiments. An example is shown in Figure 1a. The bars were then poled with an electric field between 2 and 3.5 kV mm^{-1} at 100°C for 30 minutes. The field was maintained during cooling until reaching 35°C . Afterwards, the poling was reversed with the same conditions. This was repeated three times.

Crack lengths were measured before and after the poling step and subsequent poling reversals with an optical micros-

cope, and the crack growth evaluated. Values here given for a given field and initial crack length correspond to the average of one set of four indents in a single bar. The longitudinal strain induced during the poling was evaluated by measuring the thickness of the bars before and after the electric treatment with a micrometer. The resonance of the modulus of the electric admittance was measured with a circuit as that described in (9). The dielectric permittivity was measured with a LCR meter.

3. RESULTS

X-ray diffraction patterns showed that the main phase in the ceramics was the tetragonal perovskite with a distortion of 1.023 and 1.011 for the 4D and 5H compositions, respectively.

The crack growth experiments showed that the cracks parallel to the field hardly grew for the 4D composition, and grew a small amount ($\sim 40 \mu\text{m}$ for an initial length of $440 \mu\text{m}$) for the 5H composition. On the other hand, cracks perpendicular to the field did grow significantly for both materials. This latter growth is illustrated in Figures 1b and c, in which one crack for a 4D specimen, with an initial length of $315 \mu\text{m}$, is shown before and after a poling treatment at 3.5 kV mm^{-1} . Results for the growth of cracks perpendicular to the electric field for three 4D and two 5H plates, for initial crack lengths of $336 \pm 14 \mu\text{m}$ and $440 \pm 11 \mu\text{m}$ for the 4D and 5H compositions, respectively, are shown as a function of the poling field in Figure 2a. All cracks correspond to indents introduced with the same load, 10 Kg, and therefore, the difference in the initial crack length for the two compositions is related to a difference in their fracture toughness, which is higher for the 4D material. There was no systematic trend in crack growth with electric field for either the 4D or the 5H composition. The crack growth was much greater for the 5H one. The same results combined with additional data for smaller initial crack lengths, are shown as a function of the initial length in Figure 2b. Crack growth increased with increasing initial crack length. Again, the crack growth was greater in the 5H specimens independently of the initial length.

The free stress longitudinal strain developed during poling for the two compositions is shown as a function of the electric field in Figure 3. The strain increased systematically with the poling field up to 3 kV mm^{-1} for both cases. However, the strain at 3.5 kV mm^{-1} was smaller than at 3 kV mm^{-1} , again for both PZT based compositions. Strains for a given field were always larger for the 4D material than for the 5H material. The corresponding values of the dielectric permittivity and compliance are shown in Figures 4a and b, respectively. For the 4D

TABLE I. RELATIVE DIELECTRIC PERMITTIVITY, k_3^T , LONGITUDINAL PIEZOELECTRIC COEFFICIENT, d_{33} , ELECTROMECHANICAL COUPLING FACTOR, k_{33} , COMPLIANCE, s_{11}^E , MECHANICAL QUALITY FACTOR, Q_m , DENSITY, ρ , AND TEMPERATURE OF THE FERRO-PARAELECTRIC TRANSITION, T_C , FOR THE TWO PZT BASED COMPOSITIONS STUDIED AS PROVIDED BY THE MANUFACTURER.

	k_3^T	d_{33} ($\times 10^{-12} \text{ C N}^{-1}$)	k_{33}	s_{11}^E ($\times 10^{-12} \text{ m}^2 \text{ N}^{-1}$)	Q_m	ρ (Kg m^{-3})	T_C ($^\circ\text{C}$)
4D	1300	315	.675	13.3	600	7600	320
5H	3400	593	.750	16.4	65	7450	195

composition, the dielectric permittivity increased when the field was increased from 2 to 2.5 kV mm⁻¹, decreased when the field was raised to 3 kV mm⁻¹, and increased again when it was further raised to 3.5 kV mm⁻¹. For the 5H material, the dielectric permittivity decreased continuously from 2 to 3 kV mm⁻¹, but increased when the field was further raised to 3.5 kV mm⁻¹. The compliance behaviour was similar for the two compositions, a continuous decrease up to 3 kV mm⁻¹, but an increase when the field was raised to 3.5 kV mm⁻¹.

Cracks oriented perpendicular to the electric field continued to grow during the subsequent polarisation reversals. Results with an electric field of 2.5 kV mm⁻¹ are shown for the two compositions in Figure 5. The former results on growth during poling are included for comparison (0 switching reversals). The crack growth during polarisation reversals was smaller than the one during poling, decreased with the number of reversals, and was negligible for the third reversal. Some chipping was observed to occur around the indents during the experiments. Neither the dielectric permittivity, the compliance or the piezoelectric parameters provided by the resonance technique changed significantly during the consecutive polarisation reversals.

Figure 2. Growth of the cracks perpendicular to the electric field during poling as a function of (a) the electric field, and (b) the initial crack length, for 3 4D and 2 5H plates.

4. DISCUSSION

The free stress longitudinal strain developed during poling clearly increased with the electric field in the range from 2 to 3 kV mm⁻¹ for the two materials, which was most probably related to an increasing percentage of 90° domains reorientation along the poling direction. The behaviour of the dielectric and elastic parameters support this explanation. The behaviour of the dielectric permittivity with field may be understood as a competition between two contributions linked to the reorientation process: an increase in permittivity because of the development of internal stresses (10), the permittivity ϵ_{33} has been shown to increase with the internal stress (11); and a decrease because of the reorientation process in itself, due to the crystal dielectric anisotropy, $\epsilon_{33} < \epsilon_{11}$ (12). For the 4D composition, internal stresses could have raised the permittivity from 2 to 2.5 kV mm⁻¹. From that value to 3 kV mm⁻¹ the crystallographic contribution could dominate. The tetragonal distortion of the perovskite was smaller for the 5H composition than for the 4D. Because of this, the internal stresses were most probably lower, and the crystallographic contribution could dominate within the whole range from 2 to 3 kV mm⁻¹. The decrease in the compliance when the field increased could also be related to the domain reorientation process, because of the crystal elastic anisotropy (12). On the other hand, the strain developed did not further increase when the field was raised to 3.5 kV mm⁻¹ for the two compositions, but decreased. Also, both the dielectric permittivity and the compliance increased, which seems to indicate that the percentage of domain reorientation was smaller than with 3 kV mm⁻¹. Microcracking is known to occur in PZT during poling at relative high electric fields (3). These microcracks are also known to reduce the effective field within the ceramic, leading to poor poling (4). Such a process could have happened in the current study, though it is not clear if the increase of the permittivity and compliance because of the lower degree of ferroelastic domain orientation would offset their decrease because of the presence of microcracks. The lower poling efficiency at 3.5 kV mm⁻¹ could also be linked to an increase of the leakage current. The

Figure 3. Free stress longitudinal strain developed during poling as a function of the electric field for the 4D and 5H materials.

strains developed were higher for the 4D composition than for the 5H, which could be expected, assuming a similar percentage of domain reorientation in both materials, because of the much higher tetragonal distortion of the perovskite for the former composition.

Therefore, a range of free stress longitudinal strain, from ~ 2 to 6×10^{-3} for the 4D material, and from ~ 0.5 to 2.5×10^{-3} for the 5H material, were developed as a result of an increasing percentage of 90° domain reorientation within each type. This produced different amounts of crack growth. The results for a given material show that there is no correlation between the 90° domain reorientation induced strain and the crack growth. Furthermore, the crack growth was much larger for the 5H composition in spite of the lower strains developed as compared to the 4D. These results clearly indicate that the previously proposed growth mechanism based on the strain mismatch between the untransformed crack flanks and the rest of the material does not hold. However, the reduction of the field within the crack flanks must occur, and it should lead to a mismatch, and therefore to a tensile stress at the crack faces. This stress can easily be evaluated for a simple case: a spherical untransformed region around the crack, completely embraced by the transformed ceramic. The tensile stress developed in the poling direction within the untransformed region, σ , has been shown to be given by (10):

$$\sigma = 2G(1 - \beta)\epsilon \quad [1]$$

where G stands for the shear modulus, ~ 69 for the 4D, and ~ 61 for the 5H (13), β is a function of the Poisson's ratio, which takes the value ~ 0.286 for PZT, and ϵ the free stress longitudinal strain, between 0.5 and 6×10^{-3} in our experiments. The stress generated is estimated to be between 50 and 500 MPa. Much lower values have been shown to be enough to cause 90° domain reorientation in the compositions investigated here (14). Therefore, the strain mismatch is most probably not built up in spite of the effective reduction of the electric field, because of the stress induced reorientation of domains.

We propose a mechanism for the crack growth based on the electric field intensification at the tip of the crack. This generates a stress gradient at the tip of the crack because of the piezoelectric effect, which makes the crack grow. Our results are consistent with this mechanism as the piezoelectric coefficient for the 5H material is nearly two times that of the 4D material (see Table I). Then, the stress gradient should be greater for the 5H than for the 4D material, and the crack growth greater for the 5H than for the 4D material, as observed.

Crack growth during the subsequent polarisation reversals cannot be due to the same mechanism. The d_{31} piezoelectric coefficient has not been observed to increase during subsequent reversals, and therefore, the electrically induced stress gradient should not increase either. Cao and Evans proposed a wedging mechanism, caused by localised 90° domain reorientation in contact points just behind the crack tip, to explain the observed continuous crack growth during electric cycling (6). However, we did not observe continuous crack growth but only a limited growth during the two first reversals. The observation of chipping around the indents during the reversal experiments seems to indicate that the plastic zone was not completely inactive during the experiments. Our data could be related to a wedging mechanism similar to the one proposed in (6), but involving the plastic zone rather than contact points along the crack profile.

Figure 4. Ceramics (a) relative dielectric permittivity, and (b) compliance, after poling as a function of the electric field for the 4D and 5H materials.

Figure 5. Growth of the cracks perpendicular to the electric field during consecutive polarisation reversals at 2.5 kV mm^{-1} for the 4D and 5H materials

5. CONCLUSIONS

Vicker's cracks in PZT based piezoceramics grew during poling when they were oriented perpendicular to the field. The growth did not show any correlation with the free stress longitudinal strain developed through 90° domain reorientation. The previously proposed mechanism of crack growth during poling based on a strain mismatch between the untransformed material at the crack flanks and the rest of the ceramic does not seem to hold. This is most probably because the developing stress induce 90° domain reorientation in the material at the crack flanks preventing the mismatch to be build up. Growth results are consistent with an electrically induced stress gradient, through the piezoelectric effect, at the tip of the crack.

Cracks so oriented did still grow during a small number of polarisation reversal, most probably because of a wedging effect related to 90° domain reorientation at the plastic zone around the indent.

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REFERENCES

1. K. Uchino. "Materials issues in design and performance of piezoelectric actuators: An Overview". *Acta mater.* 46:11 3745-3753.(1998)
2. S.R. Winzer, N. Shankar and A.P. Ritter. "Designing cofired multilayer electrostrictive actuators for reliability". *J. Am. Ceram. Soc.* 72:12 2246-2257. (1989)
3. E.C. Subbarao, V. Srikanth. W. Cao and L.E. Cross. "Domain switching and microcracking during poling of lead zirconate titanate ceramics". *Ferroelectrics* 145 271-281.(1993)
4. Q. Jiang, E.C. Subbarao and L.E. Cross. "Grain size dependence of electric fatigue behavior of hot pressed PLZT ferroelectric ceramics". *Acta metall. mater.* 42:11 3687-3694.(1994)
5. J. Mendiola, C. Alemany, L. Pardo and A. González. "Poling reversal effects on piezoelectricity of calcium modified lead titanate ceramic". *Ferroelectrics*, 94 209-214.(1989)
6. H. Cao and A.G. Evans. "Electric-field-induced fatigue crack growth in piezoelectrics". *J. Am. Ceram. Soc.* 77:7, 1783-1786.(1994)
7. C.S. Lynch, L. Chen, Z. Suo and R. McMeeking. "Crack growth in ferroelectric ceramics driven by cyclic polarization switching". *J. Intelligent Materials Systems and Structures* 6 191-198.(1995)
8. G.A. Schneider, H. Weitzing and B. Zickgraf. "Crack growth in ferroelectric ceramics and actuators under mechanical and electrical loading". *Fracture Mechanics of Ceramics*, 12, 149-160.(1996)
9. see for instance, "Chapter 3. Measurement techniques. C. Piezoelectric measurements", in . "Piezoelectric ceramics". B. Jaffe, W.R. Cook and H. Jaffe. Academic Press. London and New York 28-34.(1971)
10. F. Kroupa, K. Nejezchleb and I. Saxl. "Anisotropy of internal stresses in poled PZT ceramics". *Ferroelectrics* 88 123-137.(1988)
11. W.R. Buessem, L.E. Cross and A. Goswami. "Phenomenological theory of high permittivity in fine-grained barium titanate". *J. Am. Ceram. Soc.* 49:1 33-36. (1966)
12. N. Uchida and T. Ikeda. "Temperature and bias characteristics of Pb(Zr,Ti)O₃ families ceramics". *Jpn. J. Appl. Phys.* 4:11 867-880.(1965)
13. estimated from the Young's modulus, measured by compression test.
14. J.M. Calderón Moreno, F. Guiu, M. Meredith, M.J. Reece, N. McN. Alford and S.J. Penn. "Anisotropic and cyclic mechanical properties of piezoelectrics - compression testing". *J. European Ceram. Soc.* 19 1321-1324.(1999)