PERGAMON

International Journal of Solids and Structures 38 (2001) 2203–2211

INTERNATIONAL JOURNAL OF SOLIDS and STRUCTURES

www.elsevier.com/locate/ijsolstr

Critical role of domain switching on the fracture toughness of poled ferroelectrics

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Received 2 August 1999; in revised form 11 November 1999

Abstract

Domain polarization switch near the tip of a flaw plays a critical role on the fracture of poled ferroelectric ceramics under mechanical loading. Small scale 90° switching model is adopted with domain switch based on the switching released work. The model focuses on the influence on the stress intensity factor by 90° switching. Different poling directions are explored. The case of out-of-plane poling is analyzed in detail, whose result indicates appreciable shielding on the SIF for crack growing in steady state. This prediction is supported qualitatively by Vickers indents data of polycrystalline and single-crystalline ferroelectrics in which the differences in isotropic and anisotropic indentation tests can be explained qualitatively. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Domain switching; Fracture toughness; Poled ferroelectrics

1. Introduction

Ferroelectric ceramics are featured by large switching strain and low fracture toughness. The incompatible strain during domain switching may cause internal stress as high as hundreds of mega Pascals, whereas the fracture toughness assumes a typical value of $1\,\text{MPa}\sqrt{m}$. Therefore, fracture may occur from a flaw of a few microns. In fact, actuators made by ferroelectrics are often found to crack around the edges of internal electrodes. Fracture toughness anisotropy for poled ferroelectrics was extensively reported in the literature (Mehta and Virkar, 1990; Tobin and Pak, 1993; Singh and Wang, 1995) through Vickers indentation. Park and Sun (1995) performed compact tension tests under combined mechanical and electrical loading and found that the apparent fracture toughness varied asymmetrically for poled ferroelectrics under positive and negative electric fields.

The analysis for the fracture of ferroelectrics under electrical and/or mechanical loading becomes a focus point of solid mechanics. Considering the nonlinear effect, Yang and Suo (1994) modeled the electrostrictive material and derived the stress intensity factor on the flaws around the electrode edge under electric loading. Lynch et al. (1995) provided a preliminary explanation for the cracking in relaxor ferroelectrics.

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PII: S0020-7683(00)00162-1

Gao et al. (1997) and Fulton and Gao (1997) proposed a strip saturation model to investigate the effect of electric yielding.

Ferroelectrics exhibit strong nonlinear and hysteresis behaviors at a large field strength (Lynch et al., 1995; Lynch, 1998). Domain switching is the source for the hysteresis loop between the polarization and the electric field, or for the butterfly loops between the strain and the electric field. Nonlinear effect dominates in the vicinity of an internal flaw. The stress and electric fields around the flaw attempt to reorient the domains. Constrained by the unswitched material outside, the stress distribution near the flaw is altered. It is the variation of the stress intensity factor at the crack tip that dictates the apparent fracture of ferroelectrics. Pursuing this approach, the case of in-plane poling was explored by Yang and Zhu (1998). Zhu and Yang (1999) used the same idea to predict the fatigue crack growth under alternating electric field. The present work explores the effect of poling directions to the variation of the stress intensity factor at the crack tip. The theory is used to explain Vickers indentation data for PZT-5 polycrystals and for PLZT single crystals.

2. Small scale domain switching

Two types of polarization switches exist. Switch of 180° causes little strain, and is primarily activated by the electric field. Switch of 90°, however, delivers a sizeable strain of fixed amount and orientation. In the presence of electric and mechanical fields, switch of 90° is activated by the combined mechanical and electrical work (Hwang et al., 1995):

$$\sigma_{ij}\Delta\varepsilon_{ij} + E_i\Delta P_i \geqslant 2P_sE_c. \tag{1}$$

In Eq. (1), σ_{ij} and $\Delta\varepsilon_{ij}$ are the stress and the switching strain tensor, E_i and ΔP_i are the electric field and the polarization switch vectors, P_s the magnitude of the spontaneous polarization, and E_c the coercive field. The right-hand side of Eq. (1) describes an energy threshold for the polarization switch. With the absence of electric field, criterion (1) is reduced to

$$\sigma_{ij}\Delta\varepsilon_{ij} \geqslant 2P_sE_c. \tag{2}$$

Small scale domain switching consists of a practical configuration. The presence of a crack necessarily causes the stress concentration. Consequently, the zone of domain switch activated by the crack tip stress is always confined near the crack tip (Yang and Zhu, 1998).

We label the in-plane coordinates as x_1 (parallel to the crack) and x_2 (normal to the crack), and the out-of-plane coordinate as x_3. The origin of coordinates is fixed at the current crack tip. In the case of small scale switching, the geometry can be regarded as a semi-infinite crack in an otherwise infinite medium. The remote stress field is characterized by K_{app}, the applied SIF, and is given by

$$\left\{\begin{matrix}\sigma_{11}\\ \sigma_{22}\\ \sigma_{12}\end{matrix}\right\} = \frac{K_{app}}{\sqrt{2\pi r}}\cos\frac{\theta}{2}\left\{\begin{matrix}1-\sin\frac{\theta}{2}\sin\frac{3\theta}{2}\\ 1+\sin\frac{\theta}{2}\sin\frac{3\theta}{2}\\ \sin\frac{\theta}{2}\cos\frac{3\theta}{2}\end{matrix}\right\}, \tag{3}$$

where r and θ are polar coordinates centered at the crack tip.

Denote $K_{tip} = K_{app} + \Delta K$ as the stress intensity factor that governs the fracture process at the crack tip. The presence of 90° switching zone alters the near tip stress intensity factor by an amount of ΔK. The value of ΔK can be evaluated in the spirit of transformation toughening (McMeeking and Evans, 1982). For poled ferroelectrics, the toughness variation induced by switching strain can be evaluated along the boundary Γ_s of the switching zone by (Yang and Zhu, 1998)

$$\Delta K = \oint_{\Gamma_s} T_i h_i \, d\Gamma. \tag{4}$$

For an instantaneous elastic isotropic response, the amount of body force layer T_i is given by

$$T_i = 2\mu \Delta\varepsilon_{ij} n_j. \tag{5}$$

In Eq. (5), μ denotes the shear modulus, and n_j the outward normal of Γ_s. Volume conservation during domain switching is used in deriving Eq. (5). The weight function h_i in Eq. (4) denotes the SIF caused by a unit point force along the ith direction. The expressions of h_1 and h_2 are given below:

$$h_i = \frac{\tilde{h}_i}{(\kappa+1)\sqrt{2\pi r}}, \quad \begin{Bmatrix} \tilde{h}_1 \\ \tilde{h}_2 \end{Bmatrix} = \begin{Bmatrix} (1-\kappa)\cos\frac{\theta}{2} + \sin\theta \sin\frac{3\theta}{2} \\ (1+\kappa)\sin\frac{\theta}{2} - \sin\theta\cos\frac{3\theta}{2} \end{Bmatrix}. \tag{6}$$

In the case of plane strain, $\kappa = 3 - 4v$; and in the case of plane stress, $\kappa = (3 - v)/(1 + v)$, where v denotes Poisson's ratio.

3. Domain switching wake by crack tip stress field

Attention is focused on the case for crack extending in steady state. With the presence of a crack, the in-plane singular stress field may cause various domain switches of 90° near the crack tip. Domain switching wakes are formed by the activation of the crack tip stress field. Yang and Zhu (1998) studied the formation of domain switching wakes for the case of in-plane poling. A new analysis is presented herein for the case of out-of-plane poling.

Consider a specimen poled in the out-of-plane direction. Having undergone a domain switch of 90°, the out-of-plane poling axis may rotate to an in-plane polarization of any angle ω with the crack (Fig. 1). The induced domain switching strain is

$$\Delta\varepsilon_{ij} = \gamma_s \begin{bmatrix} \cos^2\omega & \sin\omega\cos\omega \\ \sin\omega\cos\omega & \sin^2\omega \end{bmatrix}, \tag{7}$$

where γ_s denotes the spontaneous strain of 90° switching. Substituting Eqs. (3) and (7) into the left-hand side of Eq. (2), one gets

$$\sigma_{ij}\Delta\varepsilon_{ij} = \frac{K_{app}\gamma_s}{\sqrt{2\pi r}} \cos\frac{\theta}{2}\left[1 - \sin\frac{\theta}{2}\sin\left(\frac{3\theta}{2} - 2\omega\right)\right]. \tag{8}$$

Polarization switching should proceed in a manner so as to release the maximum amount of that stress work. The maximization of Eq. (8) leads to the following expression of ω,

$$\omega = \frac{3\theta + \mathrm{sgn}(\theta)\pi}{4}. \tag{9}$$

The above expression implies that all domains along a radial ray from the crack tip have the same polarization switch. The value of ω jumps from $\pi/4$ to $-\pi/4$ across the crack extension line.

As a preliminary investigation on this subject, the relaxation effect is ignored. The shape of the switching zone, denoted by $R(\theta)$ hereafter, is determined by equating the maximum of the switching work to the energy threshold $2E_cP_s$. The profile is outlined by

$$\sqrt{R} = \sqrt{R_0}\cos\frac{\theta}{2}\left(1 + \left|\sin\frac{\theta}{2}\right|\right). \tag{10}$$

The size of the switching zone is scaled by

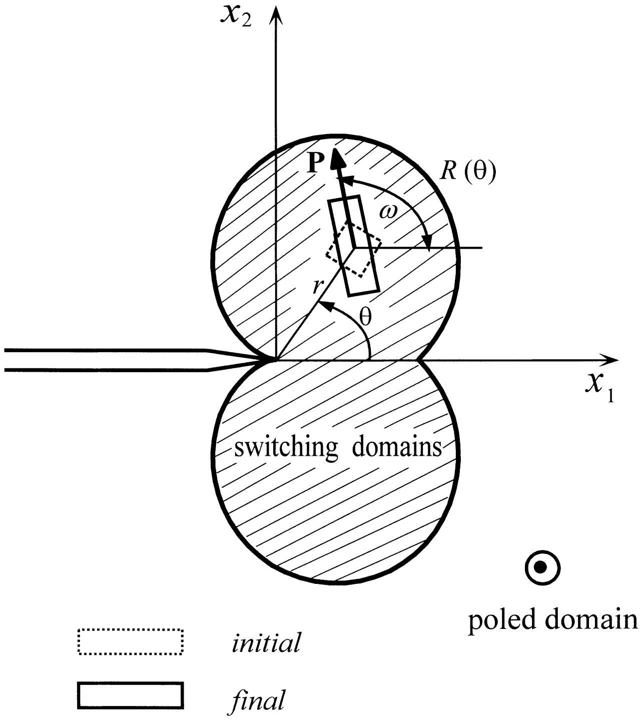

Fig. 1. Schematics of crack tip domain switch for a specimen poled out of plane.

$$R_0 = \frac{1}{8\pi}\left(\frac{K_{\rm app}\gamma_{\rm s}}{E_{\rm c}P_{\rm s}}\right)^2. \tag{11}$$

As the crack grows, switching wakes form above and below the crack, and they provide energy dissipation to toughen the ferroelectrics (McMeeking and Evans, 1982; Yang and Zhu, 1998). The height of the switching wake equals the maximum vertical position of the switching zone. For a specimen poled in the out-of-plane direction, the height of its switching wake is given by

$$H = R_0 \cos^2\frac{\overline{\theta}}{2}\left(1 + \sin\frac{\overline{\theta}}{2}\right)^2 \sin\overline{\theta}, \tag{12}$$

where the starting angle $\overline{\theta}$ for the switching wake is given by

$$3\sin 2\overline{\theta} + 4\cos\frac{3\overline{\theta}}{2} = 0. \tag{13}$$

Numerical calculations indicate that $\overline{\theta} \approx 74.84^\circ$ and $H \approx 1.5735R_0$.

4. Shielding on crack tip stress intensity factor

4.1. Shielding in a specimen poled in an in-plane direction

Yang and Zhu (1998) discussed the switch toughening under in-plane poling. We quote an expression of the apparent fracture toughness by Yang and Zhu (1998):

$$K_{\mathrm{IC}} = K_{\mathrm{intrinsic}} \Big/ \left(1 - \frac{8\mu\gamma_s^2\Omega}{(\kappa+1)P_s E_c}\right), \tag{14}$$

where $K_{\mathrm{intrinsic}}$ denotes the intrinsic fracture toughness for the sample without any domain switching and the dimensionless function Ω depends on the domain orientation. The plane strain calculation by Yang and Zhu (1998) indicated that $\Omega = 0.022$ for a polycrystalline specimen poled along the direction parallel to the crack and $\Omega = 0.044$ for poled along the direction normal to the crack. Both results were derived under the assumption that the domain polarizations are distributed uniformly within a fan between $-45°$ and $45°$ with respect to the poling axis. Yang and Zhu (1998) used the difference in Ω values to explain the fracture toughness anisotropy reported in the literature (Mehta and Virkar, 1990; Tobin and Pak, 1993; Singh and Wang, 1995).

4.2. Shielding in a specimen poled in out-of-plane direction

For a specimen poled out of plane, the shielding on the stress intensity factor can be computed by tracing various strips of infinitesimal height dy as shown in Fig. 2.

Consider a strip from y to $y + \mathrm{d}y$ within the switching wake. An anti-clockwise contour $\mathrm{d}\Gamma(y)$ is highlighted, and it is composed of two horizontal lines and one arc segment along $R(\theta)$. The switching strain within the strip can be regarded as a constant value of $\Delta\varepsilon_{ij}(y)$ so that the approach by McMeeking and Evans (1982) applies. The in-plane re-orientation of domains after the initial "lying-down" is neglected. The increment of SIF by an infinitesimal domain switch strip is

$$\mathrm{d}K(y) = \frac{2\mu\Delta\varepsilon_{ij}(y)}{\sqrt{2\pi}(\kappa+1)} \oint_{\mathrm{d}\Gamma(y)} \frac{n_j}{\sqrt{r}} \tilde{h}_i \, \mathrm{d}\Gamma. \tag{15}$$

The symmetry with respect to the crack extension line suggests the following expression of SIF by the entire switching zone:

$$\Delta K = 2\int_{y=0}^{y=H} \mathrm{d}K(y). \tag{16}$$

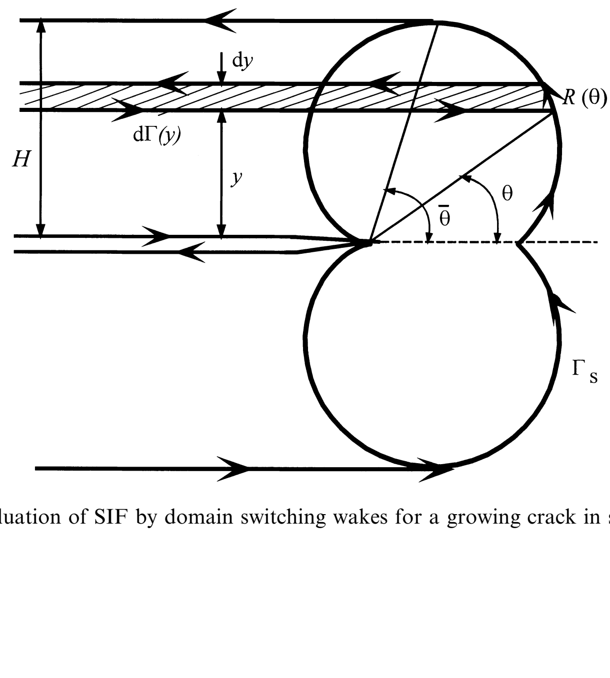

Fig. 2. Evaluation of SIF by domain switching wakes for a growing crack in steady state.

Two contributions emerge. The first one comes from the integration along all horizontal lines of $d\Gamma(y)$ with y taking all the values, strip to strip, from 0 to H. After lengthy algebraic calculations, the result is

$$\Delta K^{(1)} = \frac{-\mu\gamma_s\sqrt{R_0}}{\sqrt{2\pi}(\kappa+1)}\left\{-(3\kappa+2)\cos\frac{\overline{\theta}}{2}+\frac{1}{10}\cos\frac{5\overline{\theta}}{2}-\frac{1}{4}\sin 3\overline{\theta}+\left(\kappa-\frac{5}{8}\right)\sin 2\overline{\theta}\right.$$
$$\left.+\left(2\kappa-\frac{5}{4}\right)\sin\overline{\theta}+\frac{12}{5}+\frac{3\kappa}{2}\overline{\theta}+4\kappa\right\} \approx -\frac{2\mu\gamma_s\sqrt{R_0}}{\sqrt{2\pi}(\kappa+1)}(3.006\kappa-0.3654). \tag{17}$$

The second contribution comes from the front contour of the switching zone. That integral can be evaluated explicitly as

$$\Delta K^{(2)} = \frac{2\mu\gamma_s\sqrt{R_0}}{\sqrt{2\pi}(\kappa+1)}\left[2\kappa\sin\frac{\overline{\theta}}{2}-\frac{2\kappa+1}{3}\sin\frac{3\overline{\theta}}{2}-\frac{1}{5}\sin\frac{5\overline{\theta}}{2}\right] \approx \frac{2\mu\gamma_s\sqrt{R_0}}{\sqrt{2\pi}(\kappa+1)}(0.5983\kappa-0.3332). \tag{18}$$

The total stress intensity factor is the combination of Eqs. (17) and (18), and can be summarized as

$$\Delta K \approx -\frac{\mu}{4\pi(\kappa+1)}\frac{K_{app}\gamma_s^2}{E_c P_s}(2.4077\kappa-0.0322). \tag{19}$$

The minus sign in Eq. (19) indicates that switching strips activated by the crack tip stress field toughen the ferroelectric ceramics.

Setting the crack tip stress intensity factor, K_{tip}, to an intrinsic material parameter $K_{intrinsic}$ as proceeded by Yang and Zhu (1998), one recovers the same formula (14) for the case of out-of-plane poling. But the value of Ω is replaced by

$$\Omega = -\frac{(\kappa+1)P_s E_c}{8\mu\gamma_s^2}\frac{\Delta K}{K_{app}} = \frac{1}{16\pi}(2.4077\kappa-0.0322). \tag{20}$$

For polycrystalline PZT-5, Poisson's ratio is 1/3. Under plane strain condition, formula (20) would predict an Ω value of 0.079 for the mono-domain case. The actual domain configuration under an out-of-plane poling, however, is described by distribution of polarization vectors covering a fan of -45° to 45° from x_3 axis, see also Section 4.1. Therefore, the amplitude of switching strain should be scaled by a factor of $4/\pi\int_0^{\pi/4}\cos\varphi\, d\varphi = 2\sqrt{2}/\pi$. One may still use Eq. (14) to estimate the toughening but to scale the Ω value by a factor of $8/\pi^2$. The corrected Ω value is

$$\Omega = \frac{1}{2\pi^3}(2.4077\kappa-0.0322). \tag{21}$$

Under plane strain condition, formula (21) predicts an Ω value of 0.064, considerably higher than the one for in-plane poling.

5. Vickers indents for polycrystalline PZT-5

The material used is PZT-5 provided by the Institute of Acoustics, Chinese Academy of Sciences. The material has a tetragonal crystal structure at room temperature and an average grain size of 3 μm. Vickers indentation is carried out for $2\times4\times15$ mm^3 PZT-5 specimens. Before poling, one 4×15 mm^2 surface and one 2×15 mm^2 surface were ground and polished. Two opposing 4×15 mm^2 surfaces were sprayed with Au electrodes. The specimens are poled along the 2 mm dimension by a field of 2.5 kV/mm, well above its coercive field of 1.1 kV/mm. Microcracks normal to the poling direction are scattered along the grain boundaries. The polished surfaces were indented with a 3 kg load after removing the electrodes. The cracks emanating from the corners of the pyramid indents were measured by optical microscopy.

Since the specimens are poled through 2 mm dimension, Vickers indents are isotropic on the 4×15 mm^2 surface, and anisotropic on the 2×15 mm^2 surface. As reported in the literature (Mehta and Virkar, 1990; Tobin and Pak, 1993; Singh and Wang, 1995; among others), the indenting cracks perpendicular to the poling direction in the anisotropy plane are longer than those parallel to it. Denote $2c$ as the measured total crack length, and d as the diagonal length of the indented pyramid base. An estimate for the fracture toughness of ferroelectric ceramics is (Park and Sun, 1995)

$$K_{\mathrm{IC}} = 0.0113 \frac{d}{c^{3/2}} \sqrt{FY}, \tag{22}$$

where F is the applied indentation force, and $Y = 33$ GPa, Young's modulus of PZT-5. The diagonal of the indentation is 134 μm. In the anisotropic plane, the crack parallel to the poling direction is $c_{\parallel} = 146$ μm and the crack perpendicular to the poling direction is $c_{\perp} = 280$ μm. In the isotropic plane, all indentation cracks have the same length of about $c_{\mathrm{iso}} = 200$ μm. Thus, the apparent fracture toughness is $K_{\perp} = 0.32$ MPa$\sqrt{m}$ perpendicular to the poling direction, $K_{\parallel} = 0.85$ MPa$\sqrt{m}$ parallel to the poling direction, and $K_{\mathrm{iso}} = 0.53$ MPa$\sqrt{m}$ for the isotropic plane.

The three-dimensional nature of the indenting cracks as well as their interactions with the pre-existing microcracks makes quantitative prediction of the corresponding fracture toughness difficult. Following discussions concern only with the qualitative ranking of different apparent fracture toughnesses. Two effects may alter the apparent fracture toughness of polycrystalline PZT-5. The first effect is due to domain switching with the apparent fracture toughness predicted by Eq. (14). Since $\Omega = 0.022$ for an indenting crack normal to the poling direction, $\Omega = 0.044$ for an indenting crack parallel to the poling direction, and $\Omega = 0.064$ for an indenting crack poled out of plane, the domain switching alone would rank those fracture toughnesses as $K_{\mathrm{iso}} > K_{\parallel} > K_{\perp}$. The second effect comes from the interaction among the indenting cracks and the microcracks created during poling. These microcracks promote the extension of the indenting cracks parallel to them (as in the measurement of $K_{\perp}$), deter the extension of the indenting cracks perpendicular to them (as in the measurement of $K_{\parallel}$), and bear no effect to the extension of the indenting cracks whose fronts are along the poling direction (as in the measurement of K_{iso}). With this effect included, the ranking for apparent fracture toughnesses for ferroelectrics poled in different directions becomes $K_{\parallel} > K_{\mathrm{iso}} > K_{\perp}$ in qualitative agreement with the experimental measurements.

6. Vickers indents for ferroelectric single crystals

To exclude the complication by microcracks caused by poling, we conduct another set of Vickers indents for ferroelectric single crystals. The single crystal has a uniform lattice structure, that leads to a uniform poling. The single crystal is free of grain boundaries, which rules out the possibility of pre-existing microcracks.

The micrographs of Vickers indents on single crystal PLZT are shown in Fig. 3. The micrograph on the left refers to the case of outward poling. Domain bands perpendicular to the indenting cracks are formed, and they effectively suppress the crack extension. If one uses Eq. (22) to measure the apparent fracture toughness, its value will be high since the cracking length c in the denominator is small. That measurement agrees with the prediction in Eq. (14) since Ω assumes a large value of 0.079 for the case of a single crystal.

The micrograph on the right of Fig. 3 refers to the case of upward poling. The theoretical prediction for the apparent fracture toughness is again given by Eq. (14), where $\Omega = 0.0056$ for the prediction of $K_{\perp}$ and $\Omega = 0.062$ for the prediction of $K_{\parallel}$, see the mono-domain solution in the paper by Yang and Zhu (1998). The micrograph shows that indenting cracks under the same indenting load grow longer under upward poling than the ones under outward poling, in agreement with the theoretical prediction. Moreover, since

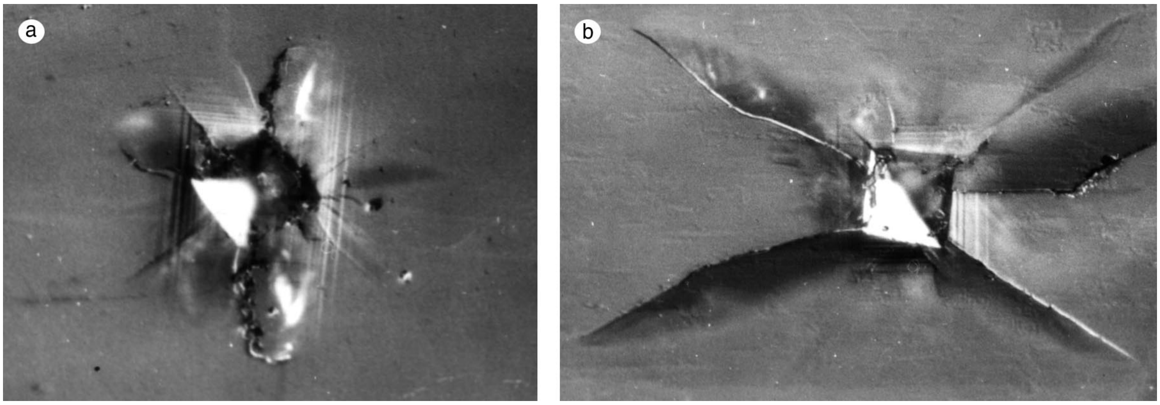

Fig. 3. Micrographs for Vickers indentation on single crystal PLZT under different polings: (a) outward poling and (b) upward poling.

the predicted value for $K_{\perp}$ is smaller than the one for $K_{\parallel}$, the indenting cracks tilt toward the direction normal to the poling axis, as would be predicted under the present model.

7. Concluding remarks

It is shown through this work that domain switching plays a critical role in the fracture and toughening of ferroelectric ceramics. As the crack grows, the domain switching wakes develop and they shield the crack tip. Understanding of this nature is similar to the case of transformation toughened structural ceramics, where the zone bearing transformation strain raises the resistance for a growing crack.

The influence of poling directions to the apparent fracture toughness is explored. In contrast to the case of in-plane poling (Yang and Zhu, 1998), the out-of-plane poling substantially raises the resistance for steady state crack growth. Consequently, large toughening can be obtained by means of out-of-plane poling, as evidenced by the Vickers indents for single crystalline ferroelectrics.

The microcracks created during poling change the fracture toughness of polycrystalline ferroelectrics. The microcracks degrades the fracture toughness for cracking perpendicular to the poling direction, shields the fracture toughness for cracking along the poling direction, and bears no effect on the fracture toughness for the case of out-of-plane poling. The fracture toughness data of polycrystal ferroelectrics should be explained by combining the effects of domain switching and microcrack interaction.

Acknowledgements

The authors would like to express their gratitude for the support given by the National Natural Science Foundation of China.

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