Fatigue crack growth in ferroelectric ceramics below the coercive field

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Perovskite-type ferroelectric ceramics have found many applications in actuators, sensors and memories. One critical limitation on their performance is due to electric fatigue, which refers to the deterioration of material properties associated with electric cycling. The degradation of electrical properties in ferroelectrics, which appears in the hysteresis loop in the form of a decrease of remanant polarization and an increase of the coercive field, is a serious concern in application [1]. Due to a strong electro-mechanical coupling effect, an electrical field may also degrade the mechanical properties of ferroelectrics. The performance of ferroelectric ceramics in smart structures is often hampered by the cracks propagating in the devices [2]. It is essential to characterize the failure behavior of ferroelectric ceramics when subjected to a cyclic electric field. Cao and Evans [3] first attacked the issue of electrical field–induced fatigue crack growth. They reported that the growth of indented cracks was governed by the magnitude of an applied electrical field E_a relative to the coercive field E_c: When $E_a \leq 0.9E_c$, there was only a minor amount of growth (about 50 μm), and then the crack arrested; when $E_a \geq 1.1E_c$, the crack continued to grow and settled into a steadily growing state. They concluded that the fatigue effect occurred only at fields above the coercive field.

For $E_a < E_c$, however, the intensified electric field near the flaw may exceed the coercive field. The electrical field concentration may cause local degradation. The analysis by Zhu and Yang [4] indicated that, for $E_a < E_c$, steady-state fatigue crack growth could be induced due to the effect of electrical field–induced domain switching at the crack tip.

We report here a detailed experimental study of electrical field–induced crack growth for ferroelectrics under an electrical field below the coercive field. In the experiment, a long-focal-length optical microscope was used to observe the crack propagation process. Optical micrographs were taken to show crack growth associated with each electrical field reversal. The reported experimental phenomenon was interpreted in terms of the domain switching model.

The material used for the experiment was PZT-5 provided by the Institute of Acoustics, Chinese Academy of Science. The material has a tetragonal crystal structure at room temperature and an average 3 μm grain size. Specimens were cut to dimensions of $4 \times 2 \times 15$ mm. Gold electrodes were sputtered onto the opposing 2×15 mm faces. The side faces were polished with 7, 5, 3.5 and 1 μm grain-sized diamond abrasive pastes. The specimens were poled at 130 °C with a poling direction along the 4 mm dimension. The specimens were poled for 0.5 h under an electrical field of 2 kV/mm. To get the coercive field, E_c, the polarization hysteresis loop was measured as a function of the electrical field. The electric displacement was monitored using the Sawyer–Tower circuit. The measured ferroelectric hysteresis loop is shown in Fig. 1. The coercive field E_c is 1100 V/mm.

The Vickers indentation technique was used to introduce the initial cracks in the electrical fatigue test. A Vickers indenter was placed in the center of the polished 4×15 mm surface with a load of 29.4N. This created a square pyramid-shaped indentation with cracks emanating from the corners. The poled ferroelectric ceramics exhibited fracture toughness anisotropy [5]. The crack perpendicular to the poling direction was much longer than the crack parallel to it. Denoting c as the measured crack length (from one corner of the indentation to the tip of the crack), the fracture toughness of the ferroelectric ceramics can be expressed as [6]

$$K_{IC} = 0.0226 \, \frac{a\sqrt{PY}}{(c+a)^{3/2}} \tag{1}$$

where Y is the Young's modulus, P is the applied indentation load and $2a$ is the length of the diagonal of the indented pyramid base.

A schematic of the experimental set-up is shown in Fig. 2. To enforce an insulated crack surface boundary condition, the specimen was immersed in a silicon oil tub, which was made of transparent and insulating Plexiglas. Cracks introduced by the Vickers indentation were subjected to a cyclic electrical field. Low-frequency rectangle wave forms were applied to the

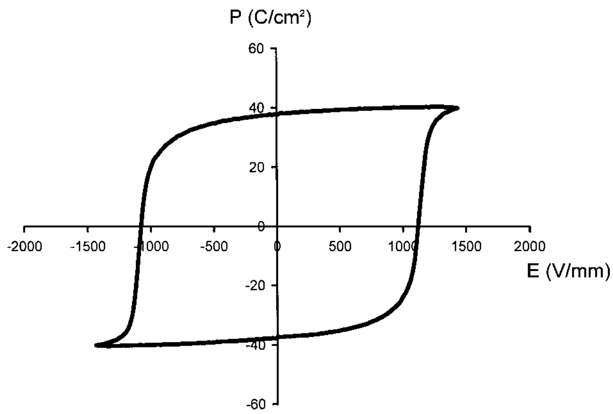

Figure 1 Hysteresis curve of electrical displacement versus electrical field.

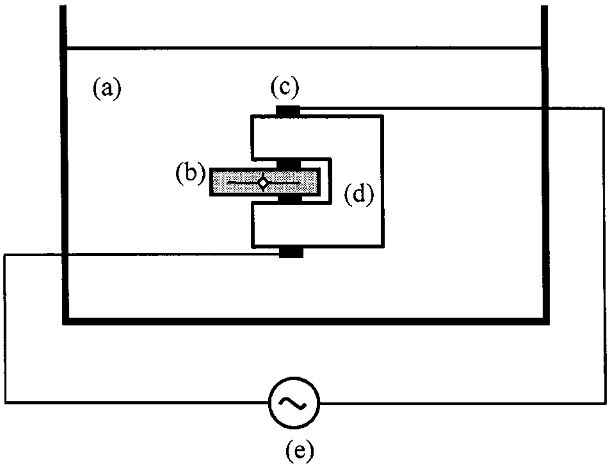

Figure 2 Schematic of the experimental set-up: (a) silicon oil; (b) specimen; (c) conducting steel rod; (d) polytetrafluoroethylene grips; and (e) high-voltage power supply.

specimen. The amplitude was 900 V/mm, which was about $0.8E_c$ and distinctly below the coercive field. A long-focal-length optical microscope was used to observe and record crack growth by means of a video imaging system. To shorten the distance between the specimen and the camera lens, the gripping device was adhered to the inner side of the oil tub.

Fig. 3a shows the optical micrograph of initial cracks emanating from the corners of the Vickers indentation. The magnification of the micrograph is 150×, and each tick corresponds to 10 μm. Crack lengths are measured from the micrograph. The crack parallel to the poling direction is 230 μm and the crack perpendicular to the poling direction is 76 μm. The diagonal of the indentation is 140 μm. Equation 1 was used to calculate the fracture toughness of the crack perpendicular to the poling direction, $K_\perp$, and the fracture toughness of the crack parallel to the poling direction, $K_{//}$. Young's modulus for PZT-5 is known to be 33 GPa. It follows that the fracture anisotropy is reflected in values of 0.30 and 0.88 MPa$\sqrt{\text{m}}$ for the cracks parallel and perpendicular to the poling direction, respectively. The anisotropy factor, defined by $K_\perp/K_{//}$, is about 3. This is in accord with the preceding measurements [7].

Fig. 3a–d show a typical crack propagation process in association with each electrical field reversal. Spots in the micrographs correspond to the pittings during polishing, and they had no significant influence on the path and the amount of crack growth. The cracks parallel to the poling direction show no significant growth. Cracks perpendicular to the poling direction, however, propagate with each electrical field reversal. Furthermore, the amount of crack growth decreases with crack extension and settles into a steadily growing state (see the curve with triangles in Fig. 4). As a departure from the experimental observation of Cao and Evans [3], we observed that each electrical field reversal drove the crack to extend a finite length even if the cyclic electrical

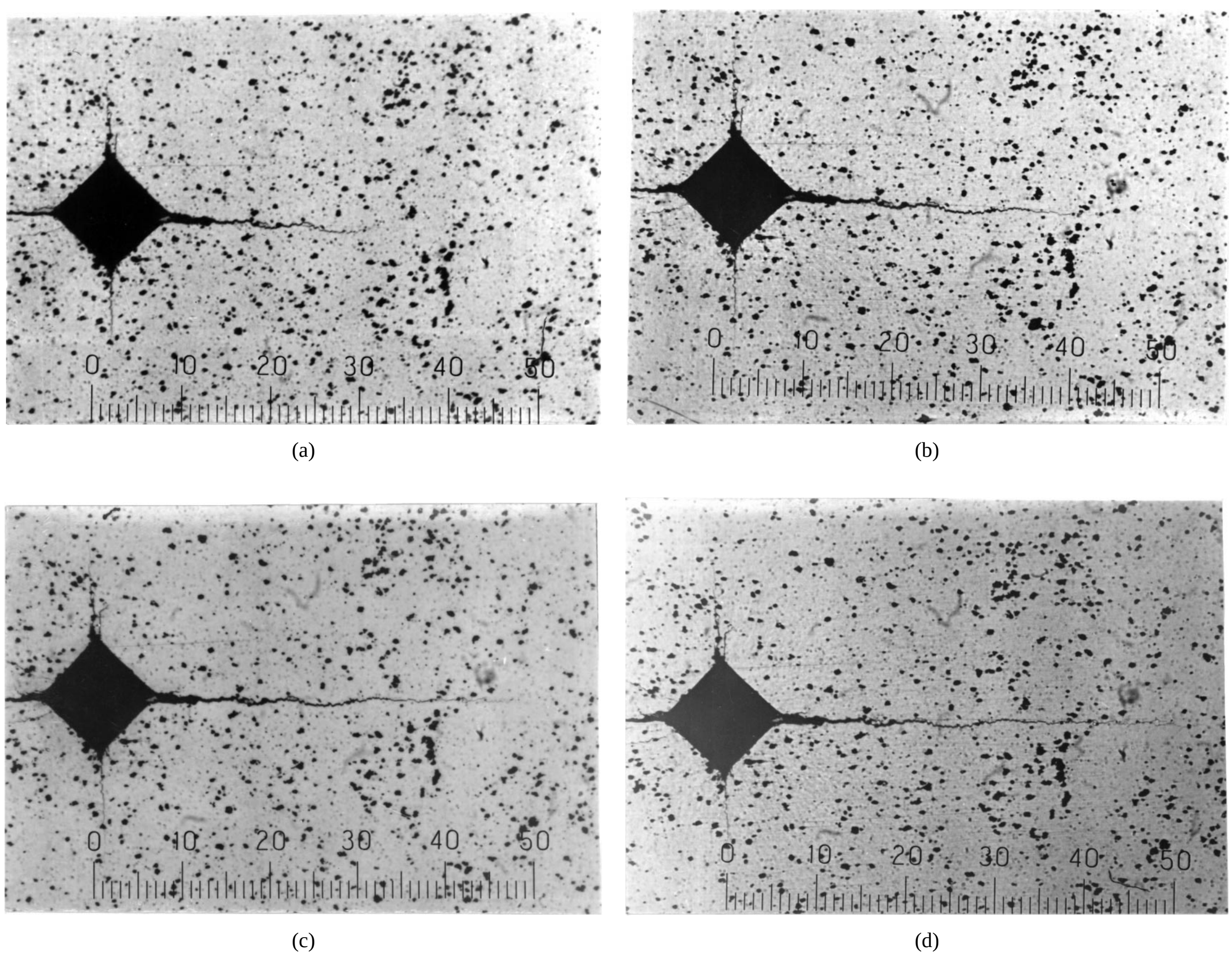

Figure 3 Optical micrographs of a crack propagation sequence in association with each electrical field reversal.

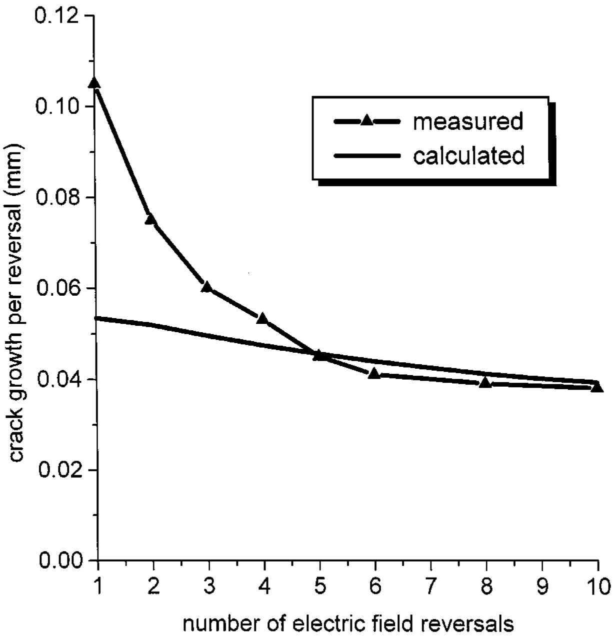

Figure 4 Increment of crack length per electrical field reversal versus number of field reversals.

field was below the coercive field, as indicated by Fig. 3a–d.

The mechanism responsible for the fatigue crack growth in ferroelectric ceramics involves electric field–induced domain switching in the vicinity of the crack tip [4, 8]. At fields below the coercive field, global domain switching will not occur. The intensified electrical field around the crack tip, however, leads to domain reorientation. A confined domain switching zone formed near the crack tip. The switched domains in the switching zone generate incompatible strain under the constraint of the surrounding unswitched material and consequently induce a singular stress field at the crack tip, which is scaled by [4]

$$K_{\text{tip}} = \Omega(\Delta a)\eta K_{\text{E}} \tag{2}$$

where K_{E} is the electric field intensity factor, $\Omega(\Delta a)$ is the function of crack extension Δa and can be determined by numerical integration and η is the following material parameter group

$$\eta = \frac{Y\gamma_{\text{s}}}{(1-\nu^2)E_{\text{c}}} \tag{3}$$

where ν is the Poisson's ratio and γ_{s} is the spontaneous strain associated with domain switching. Given the electrical field intensity factor, K_{E}, the crack will initiate if K_{tip} exceeds the intrinsic fracture toughness K_{IC} of ferroelectrics at the paraelectric phase. As the crack grows, $K_{\text{tip}}(\Delta a)$ declines monotonically. The crack arrests at $K_{\text{tip}}(\Delta a) = K_{\text{IC}}$, which determines the amount of crack increment. Upon reversal of the electrical field, domain switching is re-activated and the crack re-initiates. Thus, the crack will propagate in a repeated mode of initiation, growth, arrest and re-initiation under an alternating electrical field. The solid line in Fig. 4 gives the theoretical predication of crack increment in each electrical field reversal. In calculation, the intrinsic fracture toughness is determined by the empirical relationship given by Zhang and Raj [7]

$$K_{\text{IC}}^2 = \sqrt{2}K_{//}K_{\perp} \tag{4}$$

The electric field intensity factor is given by Suo [9]

$$K_{\text{E}} = E_{\text{a}}\sqrt{H} \tag{5}$$

where H is the width of the specimen. The Poisson's ratio is taken to be 0.3 and spontaneous strain is 0.007. In Fig. 4, the predicted crack increment shows good agreement with the experimental data as the cracking decelerates to a steadily growing state. The initial discrepancy between measured data and theoretical calculation may be caused by the three-dimensional configuration of the initial indentation crack and/or by ignoring the indentation-induced residual stress effect.

We found significant fatigue crack growth in ferroelectric ceramics under an alternating electrical field below the coercive field. Optical micrographs indicated that cracks propagated under each electrical field reversal. The origin of fatigue crack growth was attributed to the cyclic stress field induced by the repeated domain switching at the crack tip.

Acknowledgment

The authors are grateful to the support of the National Natural Foundation of China.

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Received 10 September 1998
and accepted 11 February 1999