Chapter 5

CRACKING IN FERROELECTRIC AND ANTIFERROELECTRIC CERAMIC MULTILAYER ACTUATORS

5.1 THEORETICAL APPROACHES

Although ferroelectric and antiferroelectric ceramics have different responses to the applied electric field (Fig. 1.5 and Fig. 1.6), as far as actuator reliability is concerned, they have a common property, *i.e.*, when the applied electric field reaches a certain high level, a small increase of the applied electric field results in a substantial increase of the electric displacement and the electric field induced strains. This is a main cause of the failure in a ferroelectric or antiferroelectric multilayer actuator. Yet it has not been studied. The difficulty arises from hysteresis and multiaxial loading. The strain and electric displacement depend not only on the states of loading, but also on the loading history. Generally, there are no constitutive laws available for either ferroelectric (poled or unpoled) or antiferroelectric ceramics. In this section, we propose a simple approach to an initially unpoled ferroelectric or anti-ferroelectric ceramic's dielectric property and field-induced strains at constant temperatures.

Dielectric Response

Without losing its essential feature, we idealize the nonlinear *D-E* relation on first loading (poling) of an initially unpoled ferroelectric or anti-ferroelectric ceramic under no stress in the following two forms for different purposes.

First, we write the *D-E* relation as

$$E(D) = \begin{cases} D/\varepsilon & D < D_1 \\ E_s & D_1 < D < D_2, \\ E_s + (D - D_2)/\varepsilon & D > D_2 \end{cases} \tag{5.1.1}$$

where E_s is the electric field at which the jump in the electric displacement occurs, it may be thought as the coercive field. We use this form to demonstrate the analytical electric field solution. Figure 5.1a plots this relation in the dashed line.

However, the above relation can not be conveniently used in finite element studies. Instead we use the formula,

$$\frac{D}{\varepsilon E_s} = \frac{f(E)}{\varepsilon E_s} = \frac{D_2}{2}\left\{\tanh\left[\alpha\left(\frac{E}{E_s} - 1\right)\right] + \tanh(\alpha)\right\} + \frac{E}{E_s}, \qquad (5.1.2)$$

in our finite element simulation. It is plotted in Fig. 5.1a in solid line. Ignoring the stress contribution to the electric field, we can solve the electric field distributions with nonlinear dielectric responses and finite actuator geometry using the nonlinear finite element program discussed in Chapter 4 by only interpolating and solving the electric potential ϕ.

Field Induced Strains

As majorities of the field-induced strains are due to phase transition, thus the step-like field induced strains (Fig. 5.1b) give us qualitatively physical insights. It states that, without stress, when the magnitude of the applied electric field exceeds a characteristic value, E_S, it induces not only a longitudinal tensile strain, γ_L,

$$\gamma_L = \begin{cases} \gamma_s, & |E| > E_s \\ 0, & |E| < E_s \end{cases}. \qquad (5.1.3a)$$

but also transverse strains, γ_T. The key difference between ferroelectric and antiferroelectric ceramics in this model is the sign of the transverse strains. In other words, under an applied electric field, a ferroelectric ceramic contracts transversely but an antiferroelectric ceramic elongats transversely. Write,

$$\gamma_T = \begin{cases} -q\gamma_s, & |E| > E_s \\ 0, & |E| < E_s \end{cases} \tag{5.1.3b}$$

where q is a material constant analogous to the Poisson's ratio in elasticity. It is *positive* for ferroelectric ceramics and *negative* for anti-ferroelectric ceramics. The characteristic strain, γ_s, and electric field level, E_s, necessary to induce strains play the most important roles in determining the cracking in ferroelectric and antiferroelectric ceramic multilayer actuators.

If we further assume that the ferroelectric ceramic is elastic when its polar directions in the domains are fixed by the applied electric field and take the total strains in ferroelectric ceramic to be the superposition of the elastic and field induced strains, we can write the following stress-strain relation

$$\gamma_{ij} = \left[(1+\nu)\sigma_{ij} - \nu\sigma_{mm}\delta_{ij}\right] / Y + \gamma_{ij}^{0}(E); \tag{5.1.4}$$

where $\gamma_{ij}^{0}(E)$ is the field induced strains. The relation (5.1.4) along with the equations (3.1.1)-(3.1.3) forms a standard elastic residual strain problem. After the electric field potential ϕ has been solved independently of the stress from the nonlinear finite element analysis, $\gamma_{ij}^{0}(E)$ is known, thus a linear finite element analysis can solve the stress field. Once the stress field is solved, the stress intensity factors are obtained from numerical integration (4.2.19)

5.2 ELECTRIC FIELD SOLUTION WITH NONLINEAR *D-E* RELATIONS UNDER SMALL-SCALE SATURATION CONDITIONS

Although generally one has to use the nonlinear finite element analysis to determine the electric field, under small-scale saturation conditions, electric field solutions are available for nonlinear D-E relations. We summarize the general electric field solution procedures in this section. Then we use the ideal nonlinear electric field and electric displacement relation to demonstrate the electric field solution near the end of an internal electrode.

General Solution Procedures

If we ignore the stress contribution to the electric field, we can solve the electric field for a given nonlinear electric field and electric displacement relation under small-scale saturation conditions. Noticed that all the ferroelectric ceramics are approximately linear dielectrics when the applied electric field is low, we can construct the electric field solution following the procedure by Rice (1967) under small-scale yielding conditions for strain hardening model III cracks.

Let D, E be the magnitude of electric displacement and electric field, φ be the angle between the direction of the electric field and x direction, measured positive counterclockwise. Under this notations, vectors $\boldsymbol{D}$ and $\boldsymbol{E}$ is written as,

$$\boldsymbol{D} = De^{i\varphi} = D_1 + iD_2, \quad \boldsymbol{E} = Ee^{i\varphi} = E_1 + iE_2. \tag{5.2.1}$$

Write

$$X(D) = \frac{D_1 K_E^2}{2\pi E_s}\left[2\int_D^{\infty}\frac{du}{u^2 E(u)} - \frac{1}{DE(D)}\right],$$

$$R(D) = \frac{D_1 K_E^2}{2\pi E_s DE(D)}, \tag{5.2.2}$$

where K_E is the electric field intensity factor of an asymptotic approach to the linear D-E relation. Hao *et al* (1996) pointed out that it depends on the driving voltage and the device geometry [equation (2.2.2)].

For a given electric displacement magnitude D, the position coordinates are

$$x = X(D) + R(D)cos2\varphi, \quad y = R(D)sin2\varphi. \tag{5.2.3}$$

From above expressions, one can tell that equal electric displacement magnitude lines are circles centered at ($X(D)$,0) with radius $R(D)$. The direction of the electric displacement vector at a point on the circle is along the line from the left intersection point of the circle and the x-axis point to that point, as sketched in Fig. 5.2a. The angle φ of the direction of the electric field at any point of a circle is one half the angle made with x-axis by a line from the center of the circle to that point. This means the direction of the electric field at a point on the circle is along the line from the left intersect point of the circle and the x-axis to that point. The center of the circle tends to original point as the electric displacement increases to infinity. Meanwhile, the radius of the circle vanishes.

Solution under Idealized Dielectric Responses

We now use the ***D-E*** relation in (5.1.1) to demonstrate the electric field solution. First, noticed that when $D < D_1$,

$$X(D) = \frac{K_E^2}{2\pi E_s^2}\left[1 - 2\frac{D_1}{D_2} + \frac{2D_1^2}{(D_1 - D_2)D_2} + \frac{2D_1^2}{(D_1 - D_2)^2}\ln\frac{D_2}{D_1}\right] = \text{constant}. \tag{5.2.4}$$

This means that centers of the equal field circles are fixed when $D < D_1$. This agrees with the linear dielectric asymptotic solution,

$$E_1 - iE_2 = \frac{K_E^2}{\sqrt{2\pi[Z - X(D)]}} \tag{5.2.5}$$

where Z = x+iy is the position point in complex notation.

When $D_1 < D < D_2$,

$$X(D) = R(D) + \frac{K_E^2}{2\pi E_s^2}\left[-2\frac{D_1}{D_2} + \frac{2D_1^2}{(D_1 - D_2)D_2} + \frac{2D_1^2}{(D_1 - D_2)^2}\ln\frac{D_2}{D_1}\right] \quad (5.2.6)$$

Noticed that first part decreases as D increases, while second part remains a constant, indicating that $X(D)$decreases when D increases, meaning that the center of the equal field circles moves to the negative direction of the x-axis.

When $D > D_2$,

$$X(D) = \frac{K_E^2}{2\pi E_s^2}\left[\frac{2D_1^2}{(D_2 - D_1)^2}\ln\frac{D}{D-(D_2 - D_1)} + 2\frac{D_1^2}{D(D_1 - D_2)}\right] - R(D) \quad (5.2.7)$$

Indicating that the equal field circles move towards the positive x-axis. Noticed when $D \to \infty$, $X(D)$ and $R(D)$ both go to zero.

This can also be shown by noticing that

$$X'(D)\begin{cases} = 0 & D < D_1 \\ < 0 & D_1 < D < D_2 \text{ and } R'(D) < 0, \\ > 0 & D > D_2 \end{cases} \quad (5.2.8)$$

which tells us the moving direction of the center of the circle as the electric displacement increases. In first stage, $D < D_1$, the centers coincide to a fixed point. The center of the circle moves to the negative x-direction in the second stage $D_1 < D < D_2$, and in the third stage, $D > D_2$, the center turns to the positive x-direction and moves to the original point when the electric displacement tends to infinity (Fig. 5.2b).

Denote A_1, A_2 to be the area of the circles on which the magnitudes of the electric displacement are D_1, D_2 respectively. The ratio of the area of the two circles is

$$A_2 / A_1 = (D_1 / D_2)^2. \tag{5.2.9}$$

If $D_1 / D_2 = 0.1$, then $A_2 / A_1 = 0.01$, means just one percent of the area in the circle of constant D_1 is the area in which the magnitude of the electric displacement is beyond D_2. We believe the area ratio plays an important role in actuator reliability issues and will study it in detail in the future work.

5.3 CRACKING DUE TO ELECTRIC FIELD INDUCED STRAINS

In this section, we discuss an analytical solution under small-scale saturation conditions in a limiting case of idealized dielectric properties. Then we use the nonlinear finite element program solving the electric field for a finite actuator geometry and with more realistic ***D-E*** relation. After the electric field is solved, we use the same mesh and a linear finite element analysis to solve the residual strain problem to obtain the stress field and the cracking conditions in both ferroelectrics and antiferroelectrics.

Analytical Solution in Limiting Case $D_2 \to \infty$

Under plane strain assumptions, the electric field-induced step-like strains can be written as,

$$\gamma_{ij}^{0}(E) = \begin{cases} \gamma_s\left[(1+q)\hat{E}_i\hat{E}_j - (1+\nu)q\delta_{ij}\right], & |E| > E_s \\ 0, & |E| < E_s \end{cases} \tag{5.3.1}$$

with $\hat{E}_i = E_i / E$, and E is the magnitude of the electric field, which is defined as $E = \sqrt{E_i E_i}$.

In the limiting case, when D_2 / D_1 equals to infinity, the magnitude of electric field remains constant E_s when the magnitude of the electric displacement surpasses the value of D_1. We say the electric field intensity is *saturated.* Figure 5.2b illustrates the electric field of this ideal case. It can be shown from (5.2.6) that the left intersecting point of the circles and x-axis approaches to original point. At the same time, from (5.2.2), the circle corresponding to D_2 vanishes.

As shown in Fig. 5.2b, the electric field saturates inside the cylinder with radius r_s, which is defined in (2.2.3). It centered a distance r_s ahead of the electrode, intersected the x-axis at the electrode edge, *i.e.*, original point. Inside the circle, the magnitude of the electric field intensity is E_s. It takes the same direction as the electric displacement, which is along the rays emanated from the electrode tip. Thus, at any point a distance R from the electrode edge with the angle φ made with the positive x-axis (Fig. 5.3), the electric displacement solution is written as (see Appendix A for details)

$$\begin{bmatrix} D_x \\ D_y \end{bmatrix} = \frac{2D_s r_s}{R} \begin{bmatrix} \cos\varphi \\ \sin\varphi \end{bmatrix}. \qquad (5.3.2a)$$

where $D_s = D_1 = \varepsilon E_s$. Outside the circle the ceramic is linearly dielectric. The electric field solution (5.2.5) is still valid outside the cylinder, *i.e.*, for a point a distance r from the center of the saturation cylinder (in this case the $(r_s,0)$ point) with the angle θ made with the positive x-axis (Fig. 5.3), the electric field solution is written as

$$\begin{bmatrix} D_x \\ D_y \end{bmatrix} = \frac{\varepsilon K_E}{\sqrt{2\pi r}} \begin{bmatrix} \cos\frac{\theta}{2} \\ \sin\frac{\theta}{2} \end{bmatrix}. \qquad (5.3.2b)$$

We solve the stress field around the electrode edge in the same way as Hao *et al.* (1996) and find the stresses inside the cylinder take the form (Appendix A),

$$\sigma_{xx} = GC\left(2\ln\frac{R}{r_s} - \beta - \cos 2\varphi\right)$$
$$\sigma_{yy} = GC\left(2\ln\frac{R}{r_s} - \beta + \cos 2\varphi\right)$$
$$\sigma_{xy} = -GC\sin 2\varphi \tag{5.3.3a}$$

The stresses outside the cylinder are

$$\sigma_{xx} = GC\left[2\ln\frac{R}{r} - \frac{2r_s}{r}\cos\theta + \frac{2r_s^2}{rR}\sin\theta\sin(\varphi+2\theta) - \frac{r_s^2}{r^2}(1+\beta)\cos 2\theta + 2\frac{r_s^3}{r^3}\cos 3\theta\right]$$
$$\sigma_{yy} = GC\left[2\ln\frac{R}{r} - \frac{2r_s}{r}\cos\theta - \frac{2r_s^2}{rR}\sin\theta\sin(\varphi+2\theta) + \frac{r_s^2}{r^2}(1+\beta)\cos 2\theta - 2\frac{r_s^3}{r^3}\cos 3\theta\right]$$
$$\sigma_{xy} = GC\left[-\frac{2r_s^2}{rR}\sin\theta\cos(\varphi+2\theta) - \frac{r_s^2}{r^2}(1+\beta)\sin 2\theta + 2\frac{r_s^3}{r^3}\sin 3\theta\right] \tag{5.3.3b}$$

where G is the shear modulus, C and β are defined as $C = \frac{\gamma_s(1+q)}{4(1-\nu)}$ and $\beta = 2 - \frac{4q(1+\nu)}{1+q}$.

Since the origin of the polar coordinate $(r,\ \theta)$ now coincides with the center of the saturated cylinder, the stresses are $\ln R$ singular near the electrode edge, different from the bounded stresses for linear dielectrics (Yang and Suo 1994).

Pick up a positive number of $q = 0.38$ for typical ferroelectric ceramics. Figure 5.4a displays the normal stress along the electrode plane. It localizes within a region scaled by r_S, and decays sharply away from the electrode edge. The magnitude of the stress scales with $Y\gamma_S$. It is compressive in a very small interval near the electrode edge. This is due to the idealization of the electric field and displacement relation. A tensile stress arises at a distance of $x = e^{\frac{\beta-1}{2}} r_s$ ahead of the electrode edge. When

$x > 2r_s$, it changes to compressive. The abrupt changes of the normal stress across the boundary are due to the step-like field-induced strain model.

We now consider a crack-like flaw with crack length a starting at the position $x = e^{\frac{\beta-1}{2}} r_s$, where the normal stress begins to be tensile. We obtained the stress intensity factor

$$\frac{K_I E_s}{Y\gamma_s K_E} = \frac{(1+q)e^{\frac{\beta-1}{4}}\sqrt{\alpha}}{4(1-\nu^2)}\left(\ln\frac{\sqrt{\alpha+1}+1}{2} - \frac{1-\sqrt{\alpha+1}}{2\left(1+\sqrt{\alpha+1}\right)}\right), \tag{5.3.4}$$

with $\alpha = a / r_s = 2\pi a(E_s / K_E)^2$. Due to the sudden sign change of normal stress at $x = 2r_s$, K_I maximizes when the crack reaches the boundary of the circle, *i.e.*, the crack length producing the maximum K_I is $a = \left(2e^{\frac{\beta-1}{2}} - 1\right)r_s$.

Figure 5.5a shows the stress intensity factor as a function of flaw size. The stress intensity factor vanishes for both a small and a large flaw, and peaks when the flaw reaches the boundary of the circle with crack length given in (5.3.5). This agrees with the cracking conditions established in Chapter 2. We find, in this limiting case, the coefficient in (2.2.4), $\Lambda = 0.147$.

As we stated earlier, the key difference between a ferroelectric ceramic and antiferroelectric ceramic is the sign of material constant q. We substitute q = -0.33 into equation (5.3.3) and plot the normal stress in the electrode plane in Fig. 5.4b. Different from the ferroelectric ceramics, larger tensile stress arises behind the electrode edge for antiferroelectrics. This suggests that cracking should happen behind the electrode edges. Figure 5.5b plots the stress intensity factors as a function of the flaw size for crack-like flaws starting behind the electrode edge where the stress becomes tensile. It has the same tendency in either ferroelectric or electrostrictive ceramics, *i.e.*, it vanishes for both small and large flaw sizes and peaks at a certain value of flaw size. This indicates that the model proposed in Chapter 2 applies to antiferroelectrics as well. The maximum value of the stress intensity factors indicates that $\Lambda = 0.198$ in this case.

Finite Element Analysis

We compute the stress field induced by a smooth saturated *D-E* relation to check the finite element program. The same tendencies of the stress distributions as in the analytical solutions were obtained with a smaller magnitude of maximum stress level in a narrower range and a smoother stress distribution with the singular compressive stress point shifts behind the electrode edge. We next take the ratio of $D_2 / D_1 = 6$, the distance from the end of an internal electrode edge to its closer actuator end as one and a half times the individual ceramic layer thickness. We use our nonlinear finite element program solving the electric field for this geometry. No surprising, if we normalize in the same way as we discussed in last chapter, we find all the field distributions under different loading levels look the same. This indicates that the model discussed in chapter 2 under small-scale saturation conditions applies to the large-scale saturation for ferroelectric and antiferroelectric ceramics as well. Thus, in the following discussions, we only plot a typical graph for all the cases. Figure 5.6 plots the typical normal stress distribution in the electrode plane in solid line. As D_2 is now finite, we can see that the stress is tensile in front of the electrode and decreases very fast along the positive x-direction. The stress is compressive immediately behind the electrode edge and then becomes tensile and decreases to zero. Tensile stress is more localized in front of the electrode edge. This indicates the possibilities of declined crack growths (so called Y-shape cracks) in a multilayer ferroelectric ceramic actuator. It agrees with the experimental observations (Furuta and Uchino 1993; Aburatani *et al.* 1994; Schneider *et al* 1994). To find the maximum stress intensity factor associated with this complicated stress field obtained from the finite element analysis is difficult. Thus we pick the maximum stress intensity factor in the electrode plane to approximate the value of Λ. Figure 5.7 plots the stress intensity factors as a function of flaw size in solid line. The maximum value normalized model I stress intensity factor indicates that $\Lambda \approx 0.075$. The number is smaller than the idealized solution because the smoother ***D-E*** relation is used and the stress decreases faster than the idealized case.

In the antiferroelectric analysis, we found that the largest tensile stress occurs at the point above and behind the electrode edge, suggesting that the crack should initiate a distance above and behind the electrode edge. Figure 5.6 plots the normal stress in the electrode plane in dashed line. It agrees with the analytical solution that the tensile stress behind the electrode is larger. Figure 5.7 plots the stress intensity factors in dashed line with the crack-like flaws starting at the point where σ_{yy} becomes tensile. The maximum value of the stress intensity factors indicates that $\Lambda \approx 0.053$.

5.4 CONCLUSION

We have demonstrated that the crack initiation condition established in Chapter 2 under small-scale saturation conditions is also valid for both ferroelectric and antiferroelectric ceramics using both a theoretical model and a computational model. These results confirm that, regardless of what kinds of ceramic is used in making the actuators, a critical individual ceramic layer thickness always exists below which cracking should not occur. The value of D_2/D_1 can be referenced as a parameter to characterize the nonlinearity of ***D-E*** relation. The dimensionless parameter Λ in all cases we have discussed is smaller than 0.2.