Application of Fracture Mechanics in Electrical/Mechanical Failures of Dielectrics

BY

LIU Guoning

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in Partial Fulfilment of the Requirement for
the Degree of Doctor of Philosophy
in the Department of Mechanical Engineering

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Liu Guoning

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To My Family
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Application of Fracture Mechanics in the Electrical/Mechanical Failures of Dielectrics

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Abstract

Theoretical and experimental study of the mechanical/electrical fracture behavior of dielectric materials, piezoelectric ceramics PZT 8, PZT 4, and polymeric material poly vinyl chloride (PVC), was made in this work.

The charge-free zone (CFZ) model proposed by Zhang et al. for the failure of conductive cracks in dielectrics, which was well verified by the experimental results of depoled piezoelectric ceramics PZT 4, was extended to predict the failure behavior of conductive cracks in piezoelectric ceramics. Piezoelectric ceramics were treated as mechanically brittle and electrically ductile materials in the charge-free zone model. The failure criterion, developed from the CFZ model, for conductive cracks in piezoelectric ceramics under mechanical and/or electrical loading has an elliptic shape in terms of the normalized electric intensity factor and the normalized stress intensity factor.

To verify the theoretical prediction from the CFZ model for piezoelectric ceramics, experiment was conducted to study the failure behavior of electrically conductive cracks (deep notches) in poled lead zirconate titanate PZT-8 ceramics. When the critical stress intensity factor was normalized by the critical stress intensity factor under purely mechanical loading and the critical electric intensity factor was normalized by the critical electric intensity factor under purely electric loading, the experimental results revealed that the failure behavior of the conductive cracks in the ceramics was described by an elliptic function of the normalized electric intensity factor versus the normalized stress intensity factor under combined mechanical and
electric loading. The experimental results verified well the theoretical predictions from the CFZ model.

The relationship between strain (deformation) and electric field (voltage) is linear when electric filed (voltage) is low. The relationship becomes nonlinear at high electric field (voltage). In order to adopt the linear constitutive equations which have simple forms, and to cover the actual applying procedure at the same time, a novel concept of secant piezoelectric constant was introduced to refine the CFZ model.

In addition, the concepts of fracture mechanics, which were successfully applied in the study of poled PZT 4, PZT 8 and depoled PZT 4, were introduced to study the electrical failure behavior of polymeric material poly vinyl chloride (PVC). Two kinds of samples, compact tensile (CT) samples and the double notched samples, were used to obtain the electrical fracture toughnesses. The electrical fracture toughnesses obtained from CT samples were higher than those obtained from the double notched samples. This showed that for polymers, electrical fracture toughnesses were not only related to the tested materials, but also related to the conditions of how the notch tips were electrically biased.
Chapter One

Research Objectives

Dielectric materials have been playing an important role in electric and microelectronic industries. Recently the study of failure behavior of dielectric materials, such as piezoelectric ceramics and polymers etc., has attracted great attention as their applications have been found in novel areas such as sensors, actuators, semi-conductor industries, and clean-energy industries. In industrial and academic applications of dielectric and piezoelectric materials, internal electrodes are usually embedded inside the materials. The embedded electrodes may naturally act as pre-conductive cracks and notches, which may lead to the failure of materials under electric and/or mechanical loading. Therefore, it is of academic merits and practical significance to study the failure behavior of conductive cracks or deep notches in dielectric and piezoelectric materials under mechanical or/and electrical loading. In particular, applying the concepts of fracture mechanics to the failure study of dielectric and piezoelectric materials under mechanical or/and electrical loading will advance the-state-of-the-art of fracture mechanics and make breakthrough in the reliability research of dielectric and piezoelectric materials under mechanical or/and electrical loading. The objectives of the PhD research include:
Chapter 1 Research Objectives

1. To experimentally investigate the failure behavior of conductive deep notches in piezoelectric ceramics under mechanical or/and electrical loading;

2. To further develop the charge-free zone model for piezoelectric ceramics under mechanical or/and electrical loading;

3. To apply the concepts of fracture mechanics to the failure of conductive deep notches in dielectric polymers under purely electric loading.

The research objectives aim at the establishment of failure criteria for conductive cracks and deep notches in dielectric and piezoelectric materials under mechanical or/and electrical loading. The study has high impact on academic research and engineering practice as well, which will provide design engineers the failure criteria. The output and methodologies developed here will find wide applications in academic community and in industries.
Chapter Two

Introduction

Dielectrics are usually nonmetallic materials like the polymeric materials and ceramics. Polymers are preferable materials in high voltage engineering owing to their excellent insulation properties. In power transmission system, high voltage is adopted in order to reduce the energy loss in transmission. However, high voltage may lead to dielectric breakdown of the insulation layers, which can cause serious consequences. Piezoelectric ceramics are a group of the most fascinating ceramics and have been attracting attentions in academic and research fields. Due to piezoelectricity, piezoelectric ceramics facilitate the transformation between mechanical energy and electrical energy. This makes them the preferable materials in smart structures, smart systems, actuators, and sensors. The most important piezoelectric materials are the ceramics consisting of crystallites of Perovskite structure, such as PZT, which have high piezoelectric property. PZT are alloys of lead oxide (PbO), zirconium oxide (ZrO₂), and titanium oxide (TiO₂). PZT ceramics have a cubic structure at temperature above the Curie point. Below the Curie point, piezoelectric ceramics undergo spontaneous polarization. Depending on the chemical compositions, PZT ceramics have either tetragonal structure or rhombohedral structure [1]. To minimize the potential energy, piezoelectric ceramics consist of electric domains, within which all electric dipoles are aligned in the same direction. The piezoelectric ceramics widely adopted in engineering are the poled ceramics, which are processed by heating to a temperature near or above the Curie point and then applying a
high electric field to align the dipoles until the temperature is cooling down to room temperature. The alignment of electric dipoles produces piezoelectricity at the macroscopic scale. The poled ceramics can be thermally depoled by heating to a temperature above the Curie point and then cooling down to room temperature without applying any external electric field. The thermally depoled ceramics have non-piezoelectricity at the macroscopic scale.

Piezoelectric ceramics could be subjected to mechanical and/or electrical loading in service. In smart systems like actuators, embedded metallic electrodes may act as conductive notches (cracks) and cause electrical failure/fracture under purely electrical loading. Similarly, metallic spikes in electric cables may be treated as conductive cracks in high voltage engineering, too. Therefore, the study of conductive crack problems in dielectrics, typically in polymers and piezoelectric ceramics, is of great interest not only in academic research, but also in industrial practice.

Recent development and advances in theoretical and experimental research on the fracture of piezoelectric ceramics have been summarized in review papers [2-3]. By applying the fracture mechanics to the study of electrical failure of poled piezoelectric PZT 4 ceramics [4], it was found that the electrical fracture toughness, which could be treated as a material constant to characterize the electrical failure property, was over ten times higher than the mechanical fracture toughness. The experimental study of electrical failure of thermally depoled PZT 4 ceramics [5], which had no macroscopic piezoelectricity, shows again that the electrical fracture toughness was a material
constant and its value was over ten times higher than the mechanical fracture toughness. In summary, electrical fracture consumes much higher energy than mechanical fracture.

The linear fracture mechanics shows that the electric field in the vicinity of a conductive crack has a $1/\sqrt{r}$ singularity [6], where $r$ denotes the distance from the crack tip. The high electric field at the crack tip may trigger charge emission from the crack tip by various mechanisms [7-8]. Based on the field limiting space charge (FLSC) model [9], a charge free zone (CFZ) model was proposed for the failure of depoled ceramics under combined mechanical and electric loading [10]. Macroscopically, the depoled ceramic completely lose the piezoelectricity and are dielectrics. In the previous CFZ model, there was no coupling between electric and mechanical fields. The CFZ model treats dielectric ceramics mechanically brittle but electrically ductile. The previous CFZ model is successful in explaining the physical reason behind the significant difference between the electrical fracture toughness and the mechanical fracture toughness. The experimental results on depoled PZT 4 ceramics verified the theoretical prediction of the CFZ model well. The CFZ model has been further developed in the present study by including the coupling between electric and mechanical fields such that the newly developed CFZ model can be applied to the failure of conductive cracks in piezoelectric ceramics. The detail of the newly developed CFZ model is described in Chapter 3. Experimentally, the failure of conductive cracks in piezoelectric PZT-8 ceramics was investigated under electric and/or mechanical loading. The experimental results verified the newly developed CFZ model, which were reported in Chapter 4.
For simplicity, linear constitutive equations are usually used to characterize the behavior of piezoelectric ceramics. It is well known that material behavior may become highly nonlinear at high electric field. Few literatures have considered the nonlinearity in analytical study and in numerical calculations so far. Piezoelectric properties are obtained by measuring the relationship between the electric field $E$ and induced deformation (strain, $\varepsilon$). The $\varepsilon - E$ curve is also called a butter curve due to its shape. The butter-fly curve typically shows high nonlinearity as the electric field increases. Relationship between $\varepsilon$ and $E$ is linear at low electric field and its slope is defined as a piezoelectric constant. The actual electric field near a conductive crack (notch) tip is very high before failure. Therefore, the nonlinear material behavior should be taken into account. Otherwise, the results would become inaccurate or unreliable. In Chapter 5, the CFZ model was further improved to consider the material nonlinearity. The improved CFZ model explains the experimental results perfectly.

The study on the electrical failure of polymers has been the focus from the beginning of the application of polymers in high voltage engineering [11]. Due to considerable amorphous regions in polymeric materials, the charge trapping phenomena are more significant [12]. Traditionally, the pin-plate method is used to characterize the electrical failure properties for polymers [13-14]. The electrodes in the pin-plate setup consist of a metallic rod and a plane electrode perpendicular to the rod. However, the failure parameter, the so called breakdown voltage $V_b$, is sensitive to the interface conditions and defects inside the material bulk and has a very vague physical meaning. Electrical fracture toughness, which was repeatedly proved to be a better parameter to characterize
the failure property for ceramics [4-6, 10], may be extended to study the failure of
copolymers. In Chapter 6, we report how to apply the fracture mechanics concept to the
failure of conductive cracks in dielectric polymers.
Chapter 2  Introduction

References:

Chapter Three

Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric Ceramics: Charge Free Zone (CFZ) Model

3.1 Introduction

Piezoelectric ceramics have become preferred materials for a wide variety of electronic and mechatronic devices due to their pronounced piezoelectric, dielectric, and pyroelectric properties. However, piezoelectric ceramics are brittle and susceptible to cracking at all scales ranging from electric domains to devices. Various defects, such as domain walls, grain boundaries, flaws and pores, impurities and inclusions, etc, exist in piezoelectric ceramics. The defects cause geometric, electric, thermal, and mechanical discontinuities and thus induce high stress and/or electric field concentrations, which may induce crack initiation, crack growth, partial discharge, and cause dielectric breakdown, fracture and failure. Due to the importance of the reliability of these devices, there has been tremendous interest in studying the fracture and failure behaviors of such materials [1-6]. Furthermore, internal electrodes have widely been adopted in electronic and electromechanical devices made of piezoelectric ceramics. These embedded electrodes may naturally function as pre-conductive cracks or notches, which may lead to the failure of such devices under electric and/or mechanical loads. When a conductive crack is loaded by an electrical field parallel to the crack, electric charges in the conductive crack surfaces
must rearrange themselves to produce an induced field that has the same magnitude as the applied one but with the opposite sign such that the electric field inside the conductive crack remains zero. As a result, the charges in the upper and lower crack surfaces near the crack tip have the same sign as shown in Fig.3.3. The charges with the same sign repel each other and then have a tendency to propagate the crack. The contour-independent $J$-integral used in fracture mechanics can also be applied to conductive cracks [7-11] and the $J$-integral result for a conductive crack under purely electrical loading is very much similar to that for a conventional crack under purely mechanical loading, thereby indicating that the concepts of fracture mechanics can be utilized in the study of the failure behavior of conductive cracks in ferroelectric ceramics.

Lynch et al. [12] carried out indentation fracture tests on electrode surfaces submerged in an electrically conducting NaCl solution and in distilled water. In both cases, tree-like damage grew from the indented electrode under the cyclic electric field. Heyer et al. [13] studied the electromechanical fracture toughness of conductive cracks in PZT-PIC ceramics. They conducted four-point bending tests on pre-notched bars, in which the poling direction was toward the jig surface and the notch was filled with NaCl solution to make the crack conducting. Wide-scattering results were obtained under a large applied electric field of $|K_e| > 50kV/m^{1/2}$, where $K_e$ is the applied electric intensity factor. The critical stress intensity factor increased as the applied electric intensity factor changed from $30kV/m^{1/2}$ to $-90kV/m^{1/2}$. When the applied electric intensity factor was relatively small, within the range of $-15kV/m^{1/2}$ to $15kV/m^{1/2}$, they could explain the experimental data.
using a domain-switching-based model. Using compact tension samples with conductive notches, Wang and Zhang [14] and Fu et al. [15] performed extensive fracture tests on thermally depoled and poled lead zirconate titanate (PZT-4) ceramics under purely electrical or mechanical loading. Their experimental results indicate that both the purely electric and mechanical fields can propagate conductive cracks (notches) and fracture the samples. Under purely electric loading, there exists a critical energy release rate at fracture, which is named the electric toughness. The electric toughness is much larger than the mechanical toughness, i.e., the critical energy release rate at fracture under purely mechanical loading. The high electric toughness was attributed to great energy dissipation around the conductive crack tip under purely electric loading, which is impossible under mechanical loading in the brittle electroceramics.

Recently, Zhang et al. [16] comprehensively investigated the failure behavior and the failure criterion of conductive cracks in thermally depoled PZT ceramics and proposed a charge-free zone (CFZ) model to understand the failure behavior of conductive cracks in dielectric ceramics under electrical and/or mechanical loading. The CFZ model treats dielectric ceramics as mechanically brittle and electrically ductile. Charge emission and charge trapping consume more energy and thus lead to a high value of the electric toughness. In the CFZ model, the local electric intensity factor has a non-zero value and consequently there is a non-zero local electric energy release rate, which contributes to the driving force to propagate the conductive crack. The merit of the CFZ model lies in the ability to apply the Griffith criterion directly to link the local energy release rate to the fracture toughness in a completely brittle manner. As a result, an explicit failure criterion results from the CFZ model to
predict the failure behavior of conductive cracks in dielectric ceramics under electrical and/or mechanical loading and the theoretical predictions agree perfectly with the experimental observations [16].

The CFZ model is based on the field limiting space charge (FLSC) model [17] and analogy with the dislocation-free zone (DFZ) model [18-23] and the tip-emission-adjusted zone (TEAZ) model [24-26] in the plastic fracture mechanics. The FLSC model was proposed by Zeller and Schneider [17], in which the charge mobility had only two extremes. If the electric field, $E$, is lower than a critical value of $E_c$, the value of the charge mobility is assumed to be zero in the dielectric material, whereas the charge mobility has a finite value when $E > E_c$. Based on the assumption used in the FLSC model, the level of the electric field remains the critical value in the space charge region, thereby allowing one to calculate the space charge distribution. The electric field at the tip of an electrically conductive crack is extremely high and theoretically approaches infinity. That is why an intensity factor of electric field strength, which is called the electric intensity factor in the present work for simplicity, is adopted to gauge the tip field. When the electric intensity factor reaches a critical value, charges could be emitted from the tip. Various emission mechanisms, such as the Schottky emission [27] and the Fowler-Nordheim emission [28], may function jointly at the tip. The emitted charges may form a charge cloud around the tip and thus shield the tip from the applied electric field. Injected charge carriers may be trapped at the trapping sites which could be the grain boundaries for poly-crystal materials, interfacial surfaces for layered structure as well as the line flaws like dislocations. Novel technique such as electrostatic force microscopy (EFM), which is actually a modified atomic force microscopy (AFM) technique, is widely used to
study the trapped charge carrier in the thin films. Experimental observations on
dielectric materials like silicon dioxide and semi-conductor materials like poly-
siliconor [32] and gallium nitride [34, 36] also projected some insights in the behavior
of charge carrier, charge carrier type [32-36]. The trapped charge carriers form
charge cloud near the crack tip area. At the onset of the failure of an electrically
conductive crack in a dielectric body, the charge cloud should reach a critical level.
In the proposed charge-free zone model, we shall investigate this critical level of the
charge cloud based on the concepts of fracture mechanics. To simplify our analysis,
we treat charges as line charges and the charge cloud as a charge strip in the present
study.

The DFZ model [18-23] and the TEAZ model [24-26] are proposed to understand the
fundamental issue of fracture, which is how a crack can propagate when it is
surrounded by a plastic zone, wherein the stresses are on the order of the yield
strength of the material. The conditions dominating the propagation of a crack have
to do with the theoretical strength of the bonds at the crack tip and not with the yield
strength of the macroscopic material. The common feature in the DFZ and TEAZ
models is the foundation that no dislocations exist in a small region around a crack
tip. We will not distinguish between the two models in the present work but rather
take advantages of the fact that there is a dislocation-free-region around a crack tip in
both models. We use the name DFZ here for simplicity. The DFZ model allows a
stress singularity at the crack tip such that there is a nonzero local energy release rate
to propagate the crack. It meanwhile takes the plastic deformation into account. In
the DFZ model, the Griffith fracture criterion is applied at the crack tip, whereas the
Orowan and Irwin fracture criterion is still valid macroscopically for applied loads.
Thus, one may express the two fracture criterions as

\[ G^l \geq G^l_C = \Gamma, \text{ at the crack tip,} \]  \hspace{1cm} (1)

\[ G^a \geq G^a_C = \Gamma + \Gamma_p, \text{ including the plastic zone,} \]  \hspace{1cm} (2)

where the superscripts "l" and "a" denote "local" and "applied", respectively, \( \Gamma \) represents the fracture toughness in a completely brittle manner and equals twice the specific surface energy at thermodynamic equilibrium, and \( \Gamma_p \) denotes the plastic work per surface area. Hereafter, the superscript "a" may be ignored when no confusion is caused. The two fracture criteria are used here to investigate the failure behavior of conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading.

As mentioned above, the CFZ model has successfully predicted the failure behavior of conductive cracks in thermally depoled PZT ceramics under electrical and/or mechanical loading [16]. The present work extends the CFZ model to piezoelectric ceramics, where the piezoelectric properties change, to some extent, the failure behavior. The CFZ model for piezoelectric ceramics is verified by systematically experimental tests on poled PZT-8 ceramics, which will be reported in Part II of this series.

3.2 Simplified piezoelectric approach
To clearly illustrate the physical picture of the proposed CFZ model for piezoelectric materials, we first describe the CFZ model with the simplified piezoelectric approach proposed by Gao et al. [29]. In the simplified piezoelectric formulation [29], the number of the independent material constants is reduced to a minimum. When the poling direction is along the positive $x_3$-direction, the simplified constitutive equations read

\[
\begin{bmatrix}
\sigma_{11} \\
\sigma_{22} \\
\sigma_{33} \\
\sigma_{23} \\
\sigma_{13} \\
\sigma_{12}
\end{bmatrix} = M \begin{bmatrix}
1 & * & 0 & 0 & 0 & 0 \\
* & 1 & 0 & 0 & 0 & 0 \\
* & 0 & * & 1 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & *
\end{bmatrix} \begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{23} \\
2\varepsilon_{13} \\
2\varepsilon_{12}
\end{bmatrix} \begin{bmatrix}
0 & 0 & -1 \\
0 & 0 & -1 \\
0 & 0 & 1 \\
0 & 1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 0
\end{bmatrix} \begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix},
\] (3)

\[
\begin{bmatrix}
D_1 \\
D_2 \\
D_3
\end{bmatrix} = \varepsilon \begin{bmatrix}
0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 \\
-1 & -1 & 1 & 0 & 0 & 0
\end{bmatrix} \begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{23} \\
2\varepsilon_{13} \\
2\varepsilon_{12}
\end{bmatrix} + \kappa \begin{bmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{bmatrix} \begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix},
\] (4)

where * means that the corresponding constant will not appear in the model, $\sigma_y$, $\varepsilon_y$, $D_i$ and $E_i$ denote stress tensor, strain tensor, electric displacement vector and electric field vector, respectively, and only three independent material constants $M$, $e$ and $\kappa$ are used to represent, on a qualitative basis, the elastic, piezoelectric and dielectric properties of the material. The equilibrium and kinematic equations are respectively given by

15
\[ \sigma_{y,j} = 0, \quad D_{i,j} = 0, \quad (5) \]

\[ \varepsilon_y = \frac{1}{2} (u_{i,j} + u_{j,i}), \quad E_i = -\varphi_{,i}, \quad (6) \]

where \( u \) and \( \varphi \) denote the elastic displacements and electric potential, respectively.

### 3.2.1. A single line charge near an electrical conductive crack

Consider a semi-infinite conductive crack parallel to the poling direction. The \((x, y)\) coordinate system is set up such that the crack tip is at its origin, the crack is located on the minus \(x\)-axis and the poling direction is along the positive \(x\)-direction. Constraining the elastic displacement along the \(x\)-direction, the non-vanishing displacement component in the \(y\)-direction is denoted by \( u(x, y) \). Rearranging the constitutive equations gives

\[ \sigma_{yy} = M u_{,y} + e \varphi_{,y}, \]
\[ \sigma_{yx} = M u_{,x} - e \varphi_{,x}, \]
\[ D_x = -e u_{,y} - \kappa \varphi_{,y}, \]
\[ D_y = e u_{,x} - \kappa \varphi_{,x}, \quad (7) \]

Since both \( u \) and \( \varphi \) are harmonic functions, the equilibrium Eq. (5) are satisfied automatically if \( u \) and \( \varphi \) are expressed by imaginary parts of analytic functions, respectively,

\[ u = \text{Im}[U(z)], \quad \varphi = \text{Im}[\Phi(z)], \quad (8) \]
where \( z = x + iy \). Then, the constitutive equations and kinematic equations can be expressed in terms of the analytic functions:

\[
\begin{align*}
\varepsilon_{yy} + i2\varepsilon_{yx} &= U'(z), \\
i(E_x - iE_y) &= -\Phi'(z),
\end{align*}
\]

\( i^{(9)} \)

\[
\begin{align*}
\sigma_{yy} + i\sigma_{yx} &= M U'(z) + e i \Phi'(z), \\
D_x - iD_y &= -e U'(z) + \kappa \Phi'(z).
\end{align*}
\]

\( i^{(10)} \)

The stress, strain, electric field strength, and electric displacement intensity factors are usually defined by

\[
\begin{align*}
K_o &= \lim_{z \to 0} \sqrt{2\pi z} \sigma_{yy}, & \quad K_s &= \lim_{z \to 0} \sqrt{2\pi z} \varepsilon_{yy}, \\
K_E &= \lim_{z \to 0} \sqrt{2\pi z} E_x, & \quad K_D &= \lim_{z \to 0} \sqrt{2\pi z} D_x,
\end{align*}
\]

\( i^{(11)} \)

which also satisfy the constitutive relationships given by Eq. (10). From the definitions of the intensity factors, we may express the mechanical and electrical fields near the crack in terms of the four intensity factors, i.e.,

\[
\begin{align*}
\sigma_{yy} &= \frac{K_o}{\sqrt{2\pi r}} \cos \frac{\theta}{2}, & \quad \sigma_{yx} &= -\frac{K_s}{\sqrt{2\pi r}} \sin \frac{\theta}{2}, \\
\varepsilon_{yy} &= \frac{K_e}{\sqrt{2\pi r}} \cos \frac{\theta}{2}, & \quad 2\varepsilon_{yx} &= -\frac{K_s}{\sqrt{2\pi r}} \sin \frac{\theta}{2}, \\
E_x &= \frac{K_E}{\sqrt{2\pi r}} \cos \frac{\theta}{2}, & \quad E_y &= \frac{K_E}{\sqrt{2\pi r}} \sin \frac{\theta}{2}, \\
D_x &= \frac{K_D}{\sqrt{2\pi r}} \cos \frac{\theta}{2}, & \quad D_y &= \frac{K_D}{\sqrt{2\pi r}} \sin \frac{\theta}{2},
\end{align*}
\]

\( i^{(12)} \)
where \( r \) is the distance from the crack tip and \( \theta \) is the polar angle. Using Eq. (12) and the J-integral

\[
J = \int \left( h_{n_1} - \sigma_y n_1 \mu_{s1} + D_i E_i \right) dT,
\]

we obtain the energy release rate

\[
G = J = \frac{1}{2} \left( K_{\sigma} + K_{E} + K_{\mu} \right).
\]

When a single line charge is located at \( z_d \) near a semi-infinite conductive crack, the conductive crack requires the boundary conditions of

\[
\sigma_{yy} = 0, \quad E_z = 0, \quad \text{for } x<0,
\]

along the crack faces. The complex potential satisfying the boundary conditions has the following form

\[
U = 0,
\]
\[
\Phi = \hat{Q} \ln \left( \sqrt{z} - \sqrt{z_d} \right) + \bar{\hat{Q}} \ln \left( \sqrt{z} + \sqrt{z_d} \right),
\]

where the overbar denotes the conjugate of a complex variable, and
\[ \hat{Q} = -iQ \text{ and } Q = \frac{q}{2\pi\kappa}, \] (18)

where \( q \) denotes the line charge per unit length. This line charge produces a stress field and an electric field, which are given by

\[ E_x - iE_y = \frac{Q}{2\sqrt{z}} \frac{\sqrt{z_d} + \sqrt{z}}{\sqrt{z + \sqrt{z}\left(\sqrt{z_d} - \sqrt{z}\right) - \sqrt{z_d}\sqrt{z}}}, \] (19a)

\[ \sigma_{yy} + i\sigma_{yx} = \epsilon(E_x - iE_y), \] (19b)

\[ D_x - iD_y = \kappa(E_x - iE_y). \] (19c)

It is interesting to note that the line electric charge does not produce any strain field. When the electric charge is located on the \( x \)-axis, Eq. (19a) is reduced to

\[ E_x - iE_y = \frac{q}{2\pi\kappa} \frac{\sqrt{x_d}}{\sqrt{z - x_d}}. \] (20)

Substituting Eqs. (20) and (19b) and (19c) into the intensity factor definition of Eq. (11) leads to

\[ K_E = -\frac{q}{\kappa\sqrt{2\pi x_d}}, \] (2.1a)

19
\[ K_a = eK_E, \quad \text{(21b)} \]

\[ K_D = \kappa K_E. \quad \text{(21c)} \]

These are the stress, electric and electric displacement intensity factors produced by the electric charge in front of the semi-infinity conductive crack, which are very usefully in the charge-free zone model proposed in the next section.

For the line charge near an electrical conductive crack, the applied tip field and the image field exert forces on the line charge. The image field is calculated by taking away the corresponding field of the same line charge in an infinite body from the field of the line charge with the crack and is given by

\[ E_x^{(i)} = \frac{q}{2\pi \kappa} \left[ \frac{\sqrt{x_d}}{\sqrt{x(x - x_d)}} - \frac{1}{x - x_d} \right] = -\frac{q}{2\pi \kappa} \frac{1}{\sqrt{x + \sqrt{x_d}}}. \quad \text{(22)} \]

Then, the image force per unit length calculated from \( f_i = qE_x^{(i)}(x = x_d) \) takes the form:

\[ f_i = -\frac{q^2}{2\pi \kappa} \frac{1}{2x_d}. \quad \text{(23)} \]

This image force always has the tendency to push the charge back towards the crack. On the other hand, the applied tip field exerts a driving force, \( f_a \), per unit length on the line charge, which is given by
\[ f_a = \frac{K_q}{\sqrt{2\pi x_a}}. \] (24)

The sign of \( f_a \) must be positive to emit a charge from the crack, thereby indicating that a positive (or negative) value of \( K_q \) will have the tendency to emit a positive (or negative) charge. Furthermore, the driving force must be larger than the image force in order to emit a charge from the tip. For a given applied \( K_q \), however, there exists a critical distance from the crack tip, as shown by \( x_0 \) in Figure 3.1. When the distance from the tip is smaller than \( x_0 \), the image force dominates, while the driving force dominates if the distance is larger than \( x_0 \). With the FLSC model, a charge moves forward in the region of \( x_1, x_2 \), as shown in Figure 3.1, because the total electric field is higher than \( E_c \) in the region. Thus, when a charge is emitted from the tip, it must be emitted to a distance larger than \( x_1 \). Then, the charge moves forward until it reaches \( x_2 \), beyond which the total field is lower than the critical level of \( E_c \) and the charge mobility becomes zero. The analysis indicates that, microscopically, a charge-free zone is formed adjacent to the tip in the charge emission process.

3.2.2. The charge-free zone model

In addition to the image force and the driving force, the interaction force between charges must be taken into account for many line charges. When more and more charges are emitted from the crack tip, these charges will entrap in the region of \( ba \), as shown in Figure 3.2, where \( ob \) denotes the CFZ size. If we define \( f(x') \) to be the line charge number distribution function, the charge number located at \( x' \) in the
interval \( dx' \) is \( f(x')dx' \). The equilibrium condition that the electrical field, \( E_x \), equals the critical value, \( E_c \), in the charge trap zone is described by

\[
\frac{K_b^s}{\sqrt{2\pi a}} + Q \int_b^a \frac{f(x')\sqrt{x'}}{\sqrt{x(x-x')}} dx' = E_c, \quad b \leq x \leq a.
\]  

(25)

The first term on the left in Eq. (25) represents the applied electric field, while the second term on the left stands for the electric field induced by the electric charges. The uniqueness relationship for a distribution \( f(x') \), which has zero value at \( b \) and \( a \), is given by [16]

\[
K_b^s = 2\sqrt{2\pi a} E_c E(\frac{a}{b}, k)/\pi,
\]

(26)

where \( E(\frac{a}{b}, k) \) is the complete elliptic integral of the second kind and \( k = \sqrt{1-b/a} \).

Thus, the solution to Eq. (25) is given as

\[
f(x') = -\frac{2E_c b}{\pi^2 Q \sqrt{a}} \frac{a-x'}{\sqrt{x'(x'-b)}} \Pi(\frac{a}{b}, k),
\]

(27)

where \( \Pi(\frac{a}{b}, n^2, k) \) is the complete elliptic integral of the third kind. The electrical charges produce an electric intensity factor, which is calculated by

\[
K_b^c = -\sqrt{2\pi Q} \int_b^a \frac{f(x')}{\sqrt{x'}} dx' = -\frac{2}{\pi} E_c \left[ aE(\frac{a}{b}, k) - \sqrt{b} F(\frac{a}{b}, k) \right],
\]

(28)
where the superscript \"i\" denotes the charges, and $F(\frac{\pi}{2}, k)$ is the complete elliptic integral of the first kind. Considering Eq. (26), Eq. (28) becomes to

$$K_E^i = (\Omega - 1)K_E^a,$$  \hspace{1cm} (29)

where

$$\Omega = \sqrt{\frac{b}{a}} F\left(\frac{\pi}{2}, \sqrt{1 - \frac{b^2}{a^2}}\right).$$ \hspace{1cm} (30)

Using Eqs. (21) and (29) gives the induced stress intensity factor and electric displacement intensity factor,

$$K_o^i = e(\Omega - 1)K_E^o,$$

$$K_D^i = \kappa(\Omega - 1)K_E^a.$$ \hspace{1cm} (31)

A local intensity factor is the sum of the applied intensity factor plus its corresponding intensity factor induced by the charges, i.e., $K' = K^o + K^i$, and thus, one has

$$K_o' = K_o^a + e(\Omega - 1)K_E^o,$$

$$K_E' = K_E^a,$$

$$K_D' = \Omega K_E^a,$$

$$K_D' = K_D^a + \kappa(\Omega - 1)K_E^a.$$ \hspace{1cm} (32)
Chapter 3 Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric Ceramics: Charge Free Zone (CFZ) Model

The applied strain intensity factor and the applied electric displacement intensity factor are usually expressed in terms of the applied stress and electric intensity factors by using the constitutive Eq. (10), i.e.,

\[
K_e^a = \frac{K_\sigma^a - eK_E^a}{M}, \quad K_D^a = \kappa \left(1 + \frac{e^2}{M\kappa}\right) K_E^a - \frac{e}{M} K_\sigma^a.
\]  \(33\)

Substituting Eq. (33) into Eq. (32) and then into Eq. (15) obtains the local energy release rate

\[
2G' = \frac{1}{M} \left( K_\sigma^a - eK_E^a \right)^2 + \kappa \left( \Omega K_E^a \right)^2.
\]  \(34\)

Under purely mechanical loading, applying the Griffith criterion to Eq. (34) and erasing \(K_E^a\) yield

\[
2\Gamma = \frac{1}{M} \left( K_\sigma^{o,c} \right)^2,
\]  \(35\)

where \(K_\sigma^{o,c}\) is the fracture toughness in terms of the critical stress intensity factor under purely mechanical loading and the subscript “C” denotes fracture or failure. Thus, the value of \(\Gamma\) can be evaluated from the experimental results under purely mechanical loading. Under purely electrical loading, the application of the Griffith criterion to Eq. (34) and erasing \(K_\sigma^a\) give
where $K_{E,C}^e$ is the electric fracture toughness in terms of the electric intensity under purely electrical loading. Since the electric intensity factor may be positive or negative, we have

$$K_{E,C}^e = \pm \sqrt{\frac{2\Gamma M}{\kappa M \Omega^2 + e^2}}.$$ (37)

The parameter, $\Omega$, can be evaluated from

$$\Omega = \sqrt{\frac{e^2}{\kappa M \left[ \left( \frac{K_{E,C}^e}{eK_{E,C}^e} \right)^2 - 1 \right]}}.$$ (38)

Once the value of $\Omega$ is known, we can calculate the ratio of $b/a$ from Eq. (30).

If the ratio of $b/a$ is assumed a constant, then the parameter $\Omega$ is a constant. Thus, the application of the Griffith criterion to Eq. (34) establishes the failure criterion for conductive cracks in dielectric ceramics under combined electrical and mechanical loading

$$\frac{1}{M} \left( \kappa_{\sigma,C}^a - eK_{E,C}^e \right)^2 + \kappa \left( \Omega K_{E,C}^e \right)^2 = 2\Gamma.$$ (39)
Using Eqs. (35) and (37), we can rewrite Eq. (39) in a dimensionless form:

\[
\left( \frac{K_{\sigma,c}^a}{K_{\sigma,c}^o} \right)^2 \mp \frac{2e}{\left( \varepsilon^2 + \kappa M \Omega^2 \right)^{1/2}} \left( \frac{K_{\sigma,c}^a}{K_{\sigma,c}^o} \right) \left( \frac{K_{\sigma,c}^o}{K_{\sigma,c}^a} \right) + \left( \frac{K_{\sigma,c}^o}{K_{\sigma,c}^a} \right)^2 = 1. \tag{40}
\]

Note that the negative sign is for positive electric loading and positive sign is for negative electric loading. Equation (40) establishes the failure criterion for conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading.

Mathematically, Eq. (40) has the form of

\[
x^2 + \eta xy + y^2 = 1 \text{ with } x = \frac{K_{\sigma,c}^a}{K_{\sigma,c}^o} \text{ (or } x = \frac{K_{\sigma,c}^o}{K_{\sigma,c}^a} \text{)}
\]

and \( y = \frac{K_{\sigma,c}^o}{K_{\sigma,c}^a} \text{ (or } y = \frac{K_{\sigma,c}^a}{K_{\sigma,c}^o} \text{)}, \)

and \( \eta = \mp 2e / \left( \varepsilon^2 + \kappa M \Omega^2 \right)^{1/2}. \)

The mathematic equation can be expressed in the standard form of an ellipse,

\[ x^2 /[2/(2 + \eta)] + y^2 /[2/(2 - \eta)] = 1, \]

where the \((\hat{x}, \hat{y})\) coordinator system is established by rotating \(45^\circ\) from the horizontal axis of the \((x, y)\) coordinate system.

The absolute value of \(\eta\) is less than two due to \(\left( 1 + \kappa M \Omega^2 / \varepsilon^2 \right)^{1/2} > 1\) and thus Eq. (40) indeed describes an ellipse in terms of the normalized applied intensity factors. In the case that the poling direction is along the positive \(x\)-direction, \(e\) is positive. Thus,
if applied electric fields are parallel to the poling direction, i.e., under positive electrical loading, \( \eta < 0 \) and the major semi-axis is located on the \( \hat{x} \)-axis, while \( \eta > 0 \) and the minor semi-axis is located on the \( \hat{x} \)-axis when applied electric fields are anti-parallel to the poling direction, i.e., under negative electrical loading. On the other hand, \( e \) is negative when the poling direction is along the negative \( x \)-direction. In this case, \( \eta < 0 \) and the major semi-axis is located on the \( \hat{x} \)-axis under negative electrical loading, i.e., when applied electric fields are parallel to the poling direction, whereas \( \eta > 0 \) and the minor semi-axis is located on the \( \hat{x} \)-axis under positive electrical loading, i.e., when applied electric fields are anti-parallel to the poling direction. For clarification and simplification, we may conclude that \( \eta < 0 \) and the major semi-axis is located on the \( \hat{x} \)-axis if applied electric fields are parallel to the poling direction and \( \eta > 0 \) and the minor semi-axis is located on the \( \hat{x} \)-axis if applied electric fields are anti-parallel to the poling direction. For dielectric materials, the piezoelectric constant, \( e \), is zero and thus, the interaction term, i.e., the second term on the left hand-side of Eq. (40) disappears, thereby reducing Eq. (40) to the failure criterion for conductive cracks in dielectric materials [16].

3.3 Stroh formalism

Following the previous overview article [1], we briefly introduce the two-dimensional theoretical results for a conductive crack in a piezoelectric material based on linear electro-elasticity. For readers' convenience, matrix and vector notations in this article are more consistent with those used in the book, Anisotropic Elasticity [30], but differ from the notations used in the previous review article [1].
3.3.1. Solutions for a conductive crack under remote uniform electrical and/or mechanical loading

In a rectangular coordinate system, $x_i$ ($i = 1, 2, 3$), the complete set of basic equations for a linear piezoelectric solid is given by Eqs. (5) and (6) and the constitutive equation:

$$\sigma_{ij} = C_{ijkl} e_{kl} - e_{ij} E_k,$$  \hspace{1cm} (41a)

$$D_k = e_{ij} E_i + \kappa_{ij} E_k,$$ \hspace{1cm} (41b)

where $C_{ijkl}$, $e_{ij}$ and $\kappa_{ij}$ stand for the elastic constants, the piezoelectric constants and the dielectric constants, respectively. The material constants have the following symmetries

$$C_{ijkl} = C_{jikl} = C_{ijlk}, \quad e_{ij} = e_{ji}, \quad \kappa_{ij} = \kappa_{ji}.$$ \hspace{1cm} (42)

Moreover, $C_{ijkl}$ and $\kappa_{ij}$ are positive definite in the sense that

$$C_{ijkl} u_{ij} = C_{ijkl} u_{ij} > 0, \quad \kappa_{ij} E_i E_j > 0,$$ \hspace{1cm} (43)

for arbitrary real nonzero $u_{ij}$ and $E_k$.
For generalized two-dimensional deformations in which the generalized displacement vector $\mathbf{u} = (u_1 \ u_2 \ u_3 \ \varphi)^T$ depends on $x_1$ and $x_2$ only, the general solution takes the form:

$$\mathbf{u} = \mathbf{A} \mathbf{f}(z) + \overline{\mathbf{A} \mathbf{f}(z)},$$  \hspace{1cm} (44)

$$\varphi = \mathbf{B} \mathbf{f}(z) + \overline{\mathbf{B} \mathbf{f}(z)},$$  \hspace{1cm} (45)

where $\mathbf{A} = (a_1 \ a_2 \ a_3 \ a_4)$ and $\mathbf{B} = (b_1 \ b_2 \ b_3 \ b_4)$ with $a_\alpha$ and $b_\alpha$ for $\alpha = 1, 2, 3, 4$ being both four dimensional eigen-vectors, $\mathbf{f}(z) = (f_1(z_1) \ f_2(z_2) \ f_3(z_3) \ f_4(z_4))^T$ is an analytic function vector, $z_\alpha = x_1 + p_\alpha x_2$, and $p_\alpha$ is a complex eigen-root with a positive imaginary part, and $\varphi$ is the generalized stress function vector such that

$$\Sigma_2 = (\sigma_{21} \ \sigma_{22} \ \sigma_{23} \ D_2)^T = \varphi_{11},$$  \hspace{1cm} (46a)

$$\Sigma_1 = (\sigma_{11} \ \sigma_{12} \ \sigma_{13} \ D_1)^T = -\varphi_{12}. $$  \hspace{1cm} (46b)

It is convenient to calculate $p_\alpha$, $a_\alpha$ and $b_\alpha$ by solving the following standard eigen-equation:

$$\begin{pmatrix} N_1 & N_2 \\ N_3 & N_1^T \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix} = p \begin{pmatrix} a \\ b \end{pmatrix}$$  \hspace{1cm} (47)
where \( \mathbf{N}_1 = -\mathbf{T}^{-1} \mathbf{R}^T, \mathbf{N}_2 = \mathbf{T}^{-1} = \mathbf{N}_2^T, \mathbf{N}_3 = \mathbf{R} \mathbf{T}^{-1} \mathbf{R}^T - \mathbf{Q} = \mathbf{N}_3^T \), and

\[
\mathbf{Q} = \begin{pmatrix}
    \mathbf{C}_{\text{klk1}} & \mathbf{e}_{\text{1li}} \\
    \mathbf{e}_{\text{1li}}^T & -\kappa_{\text{1li}}
\end{pmatrix},
\mathbf{R} = \begin{pmatrix}
    \mathbf{C}_{\text{klk2}} & \mathbf{e}_{\text{2li}} \\
    \mathbf{e}_{\text{2li}}^T & -\kappa_{\text{2li}}
\end{pmatrix},
\mathbf{T} = \begin{pmatrix}
    \mathbf{C}_{\text{klk2}} & \mathbf{e}_{\text{2li}} \\
    \mathbf{e}_{\text{2li}}^T & -\kappa_{\text{2li}}
\end{pmatrix}, \quad i, k = 1, 2, 3.
\]

The \( \mathbf{A} \) and \( \mathbf{B} \) matrices have the following relationship [1]

\[
\begin{pmatrix}
    \mathbf{B}^T & \mathbf{A}^T \\
    \mathbf{B} & \mathbf{A}
\end{pmatrix}
\begin{pmatrix}
    \mathbf{A} \\
    \mathbf{B}
\end{pmatrix} = \begin{pmatrix}
    \mathbf{I} & \mathbf{0} \\
    \mathbf{0} & \mathbf{I}
\end{pmatrix},
\]

(48)

where \( \mathbf{I} \) is a \( 4 \times 4 \) unit matrix. In addition, two matrices, \( \mathbf{Y} \) and \( \mathbf{H} \), are often used in the following analysis, which are defined by

\[
\mathbf{Y} = i \mathbf{A} \mathbf{B}^{-1},
\]

(49a)

\[
\mathbf{H} = 2 \operatorname{Re}[\mathbf{Y}].
\]

(49b)

Matrix \( \mathbf{Y} \) is a Hermitian matrix and can be partitioned into \([1, 31]\)

\[
\mathbf{Y} = \begin{pmatrix}
    \mathbf{Y}_e & \mathbf{Y}_{31} \\
    \mathbf{Y}_{13} & \mathbf{Y}_{44}
\end{pmatrix},
\]

(50)

where the upper left block, \( \mathbf{Y}_e \), is a \( 3 \times 3 \) matrix, and \( \mathbf{Y}_{44} \) is a real element. For a stable material, \( \mathbf{Y}_e \) is positive definite and \( \mathbf{Y}_{44} < 0 \) \([1, 31]\), which leads to \( \mathbf{H}_{44} < 0 \).
For conductive cracks, we may introduce a hybrid displacement vector, $\mathbf{u}$, and a hybrid stress function vector, $\phi$, as

$$
\mathbf{u} = (u_1, u_2, u_3, \phi)^T = \mathbf{I}_u \mathbf{u} + \mathbf{I}_\phi \phi , \quad (51)
$$

$$
\phi = (\phi_1, \phi_2, \phi_3, \varphi)^T = \mathbf{I}_\phi \mathbf{u} + \mathbf{I}_u \phi , \quad (52)
$$

where $\mathbf{I}_u$ and $\mathbf{I}_\phi$ are two diagonal matrices: $\mathbf{I}_u = \langle 1, 1, 1, 0 \rangle$ and $\mathbf{I}_\phi = \langle 0, 0, 0, 1 \rangle$. The general solution of the hybrid displacement and stress function vectors takes the form:

$$
\mathbf{u} = \hat{\mathbf{A}} f(x_\alpha) + \tilde{\mathbf{A}} f(x_\alpha), \quad (53)
$$

$$
\phi = \hat{\mathbf{B}} f(x_\alpha) + \tilde{\mathbf{B}} f(x_\alpha), \quad (54)
$$

where $\hat{\mathbf{A}} = \mathbf{I}_u \mathbf{A} + \mathbf{I}_\phi \mathbf{B}$, $\tilde{\mathbf{B}} = \mathbf{I}_\phi \mathbf{A} + \mathbf{I}_u \mathbf{B}$. The hybrid field variables can be obtained from

$$
\hat{\mathbf{S}}_2 = (\sigma_{21}, \sigma_{22}, \sigma_{23}, -E_1)^T = \hat{\phi}_{,1} , \quad (55a)
$$

$$
\hat{\mathbf{S}}_1 = (\sigma_{11}, \sigma_{12}, \sigma_{13}, E_2)^T = -\hat{\phi}_{,2} . \quad (55b)
$$
Similarly, one can define $\tilde{Y}$ and $\tilde{H}$ as

$$\tilde{Y} = i\tilde{A}\tilde{B}^{-1}, \quad \tilde{H} = 2\text{Re}[\tilde{Y}].$$

Matrix $\tilde{Y}$ is a positive definite Hermitian matrix, indicating $\tilde{Y}_{44} > 0$ and $\tilde{H}_{44} > 0$, which is different from the features of $Y_{44}$ and $H_{44}$.

Consider a conductive crack lying on the $x$-axis from $-a$ to $a$ in a piezoelectric material under remotely uniform electrical and/or mechanical loading. If no net free charges exist on the crack faces, the mechanical traction-free and the equal electrical potential boundary conditions require

$$\tilde{\Sigma}_2 = (\sigma_{21}, \sigma_{22}, \sigma_{23}, -E_1)^T = \tilde{\Phi}_1 = 0, \text{ for } -a < x < a.$$  

The analytic function, $f(x)$, satisfying the boundary conditions along the conductive crack faces is given by

$$f_{\alpha} = (\alpha_{s})_a \left( z_a + \sqrt{z_a^2 - a^2} \right) \frac{a^2 (\alpha_s)_a}{2 (z_a + \sqrt{z_a^2 - a^2})},$$

$$f_{\alpha,1} = \left( z_a + \sqrt{z_a^2 - a^2} \right) \frac{z_a + \sqrt{z_a^2 - a^2}}{2 \sqrt{z_a^2 - a^2}},$$

$$f_{\alpha,2} = p_{\alpha} f_{\alpha,1}, \quad \alpha = 1, 2, 3, 4 (\alpha \text{ not summed}),$$

$$f_{\alpha} = p_{\alpha} f_{\alpha,1}, \quad \alpha = 1, 2, 3, 4 (\alpha \text{ not summed}).$$
where \( \hat{a}_1 \) and \( \hat{a}_2 \) are both four dimensional vectors with the relationship of
\[
\hat{B}\hat{a}_2 + \overline{\hat{B}\hat{a}_1} = 0, \quad \text{and} \quad \hat{a}_1 \text{ is determined by the remote loading conditions,}
\]
\[
\begin{pmatrix}
\sigma_{12}^\infty & \sigma_{22}^\infty & \sigma_{32}^\infty & -E_z^\infty
\end{pmatrix}^T = \hat{B}\hat{a}_1 + \overline{\hat{B}\hat{a}_1},
\]
\[
\begin{pmatrix}
\sigma_{11}^\infty & \sigma_{21}^\infty & \sigma_{31}^\infty & E_z^\infty
\end{pmatrix}^T = -\hat{B}\langle p^\alpha \rangle \hat{a}_1 - \overline{\hat{B}\langle p^\alpha \rangle \hat{a}_1}.
\]

Because there are only seven independent remote input loads in Eq. (59), we take the null rotation around the \( x_3 \)-axis at infinity as a supplementary condition [1] by requiring
\[
A_2 \hat{a}_1 - \left(A\langle p^\alpha \rangle\right) \hat{a}_1 + A_3 \overline{\hat{a}_1} - \left(A\langle p^\alpha \rangle\right) \overline{\hat{a}_1} = 0.
\]

Note that a constant vector, \( f_0 \), can be added into the solution of \( f(x) \) in Eq. (58).

The constant vector, \( f_0 \), is related to the reference states of the generalized displacements and the generalized stress functions, but does not affect the stress and electric fields. Therefore, we may ignore the constant vector, \( f_0 \), and its associated constant generalized displacement and stress function vectors for simplicity.

We define an intensity factor vector at the right crack tip as
\[
K^* = \lim_{z \to \infty} \hat{B}\left(\sqrt{2\pi(z - a)}\right) f_1.
\]
Equation (61) gives

\[ f_n = \left( \frac{1}{\sqrt{2\pi a}} \right) \hat{B}^+ K^* \quad \text{for} \quad |z_a^*| < a, \quad (62) \]

where \( z_a^* = z_a - a \) and \( |z_a^*| \) is the absolute value of the complex variable \( z_a^* \). For \( |z_a^*| < a \), we consequently have

\[ f = \left( \frac{2\sqrt{z_a^*}}{\sqrt{2\pi}} \right) \hat{B}^+ K^*, \quad (63a) \]

\[ f_{1,2} = \left( \frac{p_a}{\sqrt{2\pi z_a^*}} \right) \hat{B}^+ K^*. \quad (63b) \]

A substitution of Eq. (58) into Eq. (61) yields

\[
K^* = \frac{\sqrt{\pi a}}{2} \begin{pmatrix}
\sigma_{13}^\infty \\
\sigma_{22}^\infty \\
-\sigma_{22}^\infty \\
E_{1}^\infty
\end{pmatrix} = \frac{1}{2} \begin{pmatrix}
K_{II} \\
K_I \\
K_{III} \\
-K_E
\end{pmatrix} = \frac{1}{2} K, \quad (64)
\]

which shows that the intensity factor vector \( K^* \) is real and its components equal half of the stress intensity factors of mode II, I, and III and the electric intensity factor. Then, the mechanical and electrical fields near the crack tip can be approximately expressed by
\[
\hat{u} = \frac{1}{\sqrt{2\pi}} \left[ \hat{A} \left( \sqrt{z_a^*} \right) \hat{B}^{-1} + \overline{\hat{A}} \left( \sqrt{z_a^*} \right) \overline{\hat{B}}^{-1} \right] \mathbf{K}, \tag{65a}
\]

\[
\hat{\Sigma}_2 = \frac{1}{2\sqrt{2\pi}} \left[ \hat{B} \left( \frac{1}{\sqrt{z_a^*}} \right) \hat{B}^{-1} + \overline{\hat{B}} \left( \frac{1}{\sqrt{z_a^*}} \right) \overline{\hat{B}}^{-1} \right] \mathbf{K}, \tag{65b}
\]

\[
\hat{\Sigma}_4 = -\frac{1}{2\sqrt{2\pi}} \left[ \hat{B} \left( \frac{p_a}{\sqrt{z_a^*}} \right) \hat{B}^{-1} + \overline{\hat{B}} \left( \frac{p_a}{\sqrt{z_a^*}} \right) \overline{\hat{B}}^{-1} \right] \mathbf{K}. \tag{65c}
\]

The generalized crack opening near the crack tip is

\[
\Delta \hat{u} = \frac{\sqrt{2r}}{\sqrt{\pi}} \hat{H} \mathbf{K}. \tag{66}
\]

From Eq. (13), we have the J-integral or the energy release rate

\[
J = \mathbf{K}^T \frac{\hat{H}}{4} \mathbf{K}. \tag{67}
\]

3.3.2. Interaction of a piezoelectric dislocation with a conductive crack

For a piezoelectric dislocation located at \( z_a^d \) in an infinite piezoelectric medium, the analytic function vector is given by
\[ f(z) = \left\langle \ln \left( z_a - z'_a \right) \right\rangle q, \quad (68) \]

where the \( q \) vector represents the feature of a generalized piezoelectric dislocation and combines the generalized Burgers vector, \( b \), and the generalized force, \( F \),

\[ q = \frac{1}{2\pi i} \left( A^T F + B^T b \right). \quad (69) \]

The generalized Burgers vector, \( b \), and the generalized force, \( F \), are defined as \( b = (b_1, b_2, b_3, \Delta \phi)^T \) and \( F = (F_1, F_2, F_3, -q)^T \), respectively, where \( b_i \) and \( F_i \) denote the component of the Burgers vector and a line force per unit length along the \( x_i \) direction, respectively, \( \Delta \phi \) is the electric potential jump, and \( q \) stands for a line charge per unit length.

When the dislocation is near a finite length conductive crack, the solution, satisfying the boundary conditions of Eq. (57) along the conductive crack faces, takes the form:

\[ f_{\rightarrow} = \left\langle \frac{1}{\sqrt{z'^2 - a^2}} \left( \frac{z_a + \sqrt{z'_a - a^2}}{z_a + \sqrt{z'_a - a^2} - z'_a - \sqrt{\left( z'_a \right)^2 - a^2}} \right) \right\rangle q + Z_{\rightarrow}^- q, \quad (70a) \]
\[ Z_{\eta,1} = \sum_{k=1}^{4} \hat{B}_{ik}^{-1} \bar{B}_{kj} \frac{1}{\sqrt{z_i^2 - a^2}} \left( \frac{a^2}{a^2 - \left( z_i + \sqrt{z_i^2 - a^2} \right) \left( z_j + \sqrt{z_j^2 - a^2} \right)} \right) \] \quad \text{(70b)}

\[ f_{\eta,2} = \left( \frac{p_{\alpha}}{\sqrt{z_{\alpha}^2 - a^2}} \left( \frac{z_{\alpha} + \sqrt{z_{\alpha}^2 - a^2}}{z_{\alpha} + \sqrt{z_{\alpha}^2 - a^2} - z_{\alpha}^d - \sqrt{z_{\alpha}^d - a^2}} \right) \right) q + Z_{\eta,2} q, \quad \text{(70c)} \]

\[ Z_{\eta,3} = \sum_{k=1}^{4} \hat{B}_{ik}^{-1} \bar{B}_{kj} \frac{p_{\alpha}}{\sqrt{z_i^2 - a^2}} \left( \frac{a^2}{a^2 - \left( z_i + \sqrt{z_i^2 - a^2} \right) \left( z_j + \sqrt{z_j^2 - a^2} \right)} \right) \] \quad \text{(70d)}

For a semi-infinite crack with the origin of the coordinate system at the crack tip, Eqs. (70a-d) are reduced to

\[ f_{\eta,1} = \left( \frac{1}{2\sqrt{z_{\alpha}} \left( \sqrt{z_{\alpha}^2 - z_{\alpha}^d} \right)} \right) q + Z_{\eta,1} q, \quad \text{(71a)} \]

\[ Z_{\eta,1} = -\sum_{k=1}^{4} \hat{B}_{ik} B_{kj} \frac{1}{2\sqrt{z_i} \left( \sqrt{z_i} + \sqrt{z_j^d} \right)} \] \quad \text{(71b)}

\[ f_{\eta,2} = \left( \frac{p_{\alpha}}{2\sqrt{z_{\alpha}} \left( \sqrt{z_{\alpha} - z_{\alpha}^d} \right)} \right) q + Z_{\eta,2} q, \quad \text{(71c)} \]
\[ Z_{\mu,\nu} = -\sum_{l=1}^{4} \hat{B}_{x} \hat{B}_{y} \frac{p_{l}}{2\sqrt{z_{l}} \left( \sqrt{z_{l}} + \sqrt{z_{l}^{2} + a^{2}} \right)}. \]  \hspace{1cm} (71d)

The intensity factor vector at the right crack tip induced by the dislocation at \( z_{a} \) is

\[ K^{*} = \lim_{z_{a} \to a} \hat{B}_{\left( \sqrt{2\pi(z_{a} - a)} \right)} f_{1} = \frac{1}{\sqrt{\pi a}} \left[ \frac{1}{z_{a} - a + \sqrt{(z_{a}^{2}) - a^{2}}} \hat{B} q + \frac{1}{\sqrt{z_{a}^{2} - a + \sqrt{(z_{a}^{2})^{2} - a^{2}}} \hat{B} q \right], \]  \hspace{1cm} (72)

for a finite conductive crack and

\[ K^{*} = \lim_{z_{a} \to 0} \hat{B}_{\left( \sqrt{2\pi a} \right)} f_{1} = -\frac{\sqrt{2\pi}}{2} \left[ \hat{B} \left( \frac{1}{\sqrt{z_{a}}} \right) q + \hat{B} \left( \frac{1}{\sqrt{z_{a}}} \right) \right], \]  \hspace{1cm} (73)

for a semi-infinite conductive crack. Equations (72) and (73) indicate that the intensity factors induced by a dislocation are real such that \( K^{*} = K/2 \) holds. As a consequence, Eqs (65-67) give also the crack tip fields and the energy release rate produced by the piezoelectric dislocation, respectively.

The image field of a piezoelectric dislocation induced by the crack is calculated by taking away the corresponding field of the same dislocation in an infinite body from the field of the dislocation with the crack. Differentiating Eq. (68) with respect to \( x_{1} \)
and \( x_2 \) and then subtracting the results correspondingly from Eqs. (70a, c) and Eqs. (71a, c) yield the analytic vectors of the image field:

\[
f_{i,i} = \left( \frac{-a^2 (z_a + z_a^d)}{\sqrt{z_a^2 - a^2} \left( z_a + \sqrt{z_a^2 - a^2} + z_a^d + \sqrt{(z_a^d)^2 - a^2} \right) \left( z_a \sqrt{(z_a^d)^2 - a^2} + z_a^d \sqrt{z_a^2 - a^2} \right)} \right) q + Z_{i,i} q,
\]

\[
f_{i,s} = \left( \frac{-a^2 p_a (z_a + z_a^d)}{\sqrt{z_a^2 - a^2} \left( z_a + \sqrt{z_a^2 - a^2} + z_a^d + \sqrt{(z_a^d)^2 - a^2} \right) \left( z_a \sqrt{(z_a^d)^2 - a^2} + z_a^d \sqrt{z_a^2 - a^2} \right)} \right) q + Z_{i,s} q,
\]

for the finite length crack, and

\[
f_{i,i} = \left( \frac{-1}{2 \sqrt{z_a} \left( z_a + \sqrt{z_a^2} \right)} \right) q + Z_{i,i} q,
\]

\[
f_{i,s} = \left( \frac{-p_a}{2 \sqrt{z_a} \left( z_a + \sqrt{z_a^2} \right)} \right) q + Z_{i,s} q,
\]

for the semi-infinite length crack.

3.3.3. A single line charge near an electrical conductive crack
For clearance and simplicity, hereafter we use $x$ and $y$ to replace $x_1$ and $x_2$, respectively. Consider a line charge located at $x_d$ on the $x$-axis in front of a semi-infinite conductive crack. For a line charge, the feature vector, $q$, takes the form:

$$q = -\frac{q}{2\pi i} \begin{pmatrix} A_{41} & A_{42} & A_{43} & A_{44} \end{pmatrix}^T = -\frac{q}{2\pi i} A_4^T,$$

(76)

where $A_4$ is the fourth row of the matrix, $A$. The electric field, $E_x$, along the $x$-axis produced by the line charge is calculated by using Eq. (71) and is given by

$$E_x = \frac{q}{2\pi \kappa_e} \frac{\sqrt{x_d}}{\sqrt{x(x-x_d)}},$$

(77)

where $\kappa_e$ denotes an effect dielectric constant, which is defined as

$$\kappa_e = \frac{i}{A_4A_4^T - A_4A_4^T}.$$

(78)

In the proposed CFZ model, we emphasize on the charge trapping near the crack tip and thus, the applied tip fields are given in terms of applied intensity factors. From Eq. (63b), we have the near tip field along the $x$-axis in terms of the electric intensity factor

$$E_x = -\frac{1}{\sqrt{2\pi \kappa}} K_E.$$

(79)
From Eq. (73), we calculate the intensity factor vector produced by the line-charge

\[
\begin{pmatrix}
K_{II} \\
K_I \\
K_{III} \\
-K_E
\end{pmatrix} = \sqrt{2\pi} \frac{q}{\lambda} \left[ \frac{\bar{B}A_1^\top - \bar{B}A_4^\top}{\sqrt{2\pi}} \right]
\]

Equation (80) can be rewritten as

\[
\begin{pmatrix}
K_{II} \\
K_I \\
K_{III} \\
K_E
\end{pmatrix} = \begin{pmatrix}
\Lambda_1 \\
\Lambda_2 \\
\Lambda_3 \\
\Lambda_4
\end{pmatrix} K_E^q = \Lambda K_E^q,
\]

where

\[
\Lambda_j = -\frac{B_j A_1^\top - B_j A_4^\top}{A_4 A_1^\top - A_4 A_4^\top}, \quad (j = 1, 2, 3), \quad \Lambda_4 = 1,
\]

and

\[
K_E^q = -\frac{q}{\kappa_\epsilon \sqrt{2\pi \lambda}}.
\]

Equation (81) shows that the line charge produces an electric intensity factor and stress intensity factors and the induced stress intensity factors are proportional to the induced electric intensity factor.
For the line charge located on the x-axis near an electrical conductive crack, the applied tip field and the image field exert forces on the line charge. The image field is calculated from Eq. (75a) to be

\[
E_s^{(i)} = -\frac{q}{2\pi \varepsilon_x} \frac{1}{\sqrt{x} \left(\sqrt{x} + \sqrt{x_d}\right)}.
\]  

(84)

Then, the image force per unit length calculated from \( f_i = qE_s^{(i)} (x = x_d) \) is given by

\[
f_i = -\frac{q^2}{2\pi \varepsilon_x} \frac{1}{2x_d}.
\]  

(85)

This image force always has the tendency to push the charge back towards the crack. On the other hand, the applied tip field exerts a driving force, \( f_a \), per unit length on the line charge, which is given by

\[
f_a = \frac{K_s q}{\sqrt{2\pi x_d}}.
\]  

(86)

Equations (85) and (86) are the same as Eqs. (23) and (24). Thus, the analysis of the charge emission will be identical to that described in Section 2.1, which will not be repeated here.

3.3.4. The charge-free zone model
Identical to Eq. (25), the equilibrium condition that the electrical field, $E_e$, equals the critical value, $E_c$, in the charge trap zone is described by

$$\frac{K_e^a}{\sqrt{2\pi}} + Q \int_b^a \frac{f(x')\sqrt{x'}}{\sqrt{x(x-x')}} \, dx' = E_c, \quad b \leq x \leq a,$$

(87)

where

$$Q = \frac{q}{2\pi\kappa_e}.$$

(88)

Equations (26-30) are also the solutions to Eq. (87). Next, using Eq. (81) and Eq. (29), we have the induced intensity factor vector

$$\mathbf{K}^I = \mathbf{K} + \mathbf{K}'$$

(89)

A local intensity factor is the sum of the applied intensity factor plus its corresponding intensity factor induced by the charges,

$$\mathbf{K}' = \mathbf{K}^a + \mathbf{K}'$$

(90)

Finally, one has

$$\mathbf{K}' = \begin{pmatrix} K_{II}' \\ K_{I}' \\ K_{III}' \\ K_e' \end{pmatrix} = \begin{pmatrix} K_{II}' + \Lambda_1(\Omega-1)K_e^a \\ K_{I}' + \Lambda_4(\Omega-1)K_e^a \\ K_{III}' + \Lambda_4(\Omega-1)K_e^a \\ \Omega K_e^a \end{pmatrix}.$$
Equation (91) shows that the local intensity factors depend on the applied intensity factors and the parameter, $\Omega$. If we assume that the ratio of $b/a$ is a constant, the $\Omega$ parameter is a constant. Using Eq. (67), we may then express the local energy release rate in terms of the local intensity factors as

$$J^l = (K^l)^T \hat{H} K^l.$$  \hfill (92) 

The application of the Griffith criterion to Eq. (92) yields

$$J^l_C = (K^l_C)^T \hat{H} K^l_C = \Gamma,$$  \hfill (93) 

where the subscript "C" is labeled to indicate the fracture equilibrium condition. In most cases, mechanical loading is applied in mode I. Under purely mechanical mode I loading, substituting Eq. (91) into Eq. (93) yields

$$G_{\sigma,C} = J^l_C = \frac{\hat{H}_m}{4} \left( K_{\sigma,C}^l \right)^2 = \Gamma,$$  \hfill (94) 

where the superscript "o" denotes "purely" mechanical or electrical loading, $K_{\sigma,C}^o = K_{I,C}^o$ and hereafter we shall use the subscript "o" to replace "I" in order to be consistent with the notation used in the simplified approach. Equation (94) indicates that the value of $\Gamma$ is identical to the mechanical toughness, $G_{\sigma,C}$. Similarly, we have the expression of the critical local $J$-integral under purely electrical loading.
\[ G_{e,c} = J_C' = \Theta \begin{pmatrix} K_{e,c}^2 \end{pmatrix}^2 = \Gamma, \quad (95a) \]

\[ \Theta = \begin{pmatrix} \Lambda_1(\Omega - 1) & \Lambda_2(\Omega - 1) & \Lambda_3(\Omega - 1) & \Omega \end{pmatrix} \begin{pmatrix} \frac{\Lambda_1(\Omega - 1)}{4} \\ \frac{\Lambda_2(\Omega - 1)}{4} \\ \frac{\Lambda_3(\Omega - 1)}{4} \\ \Omega \end{pmatrix}, \quad (95b) \]

Both \( K_{e,c}^2 \) and \( K_{e,c}^0 \) can be determined experimentally. Thus, we can determine the value of \( \Theta \),

\[ \Theta = \frac{\hat{H}_{22}}{4} \begin{pmatrix} K_{e,c}^2 \end{pmatrix}^2, \quad (96) \]

and the value of \( \Omega \) from Eq. (95b).

For a transversely isotropic piezoelectric material with its poling direction along the \( x \)-axis, the calculation results indicate that \( \Lambda_1 = \Lambda_3 = 0 \). Then, we can simplify Eq. (95b) to

\[ \Theta = \frac{1}{4} \left[ \Lambda_2^2(\Omega - 1)^2 \hat{H}_{22} + 2 \Lambda_1(\Omega - 1)\Omega \hat{H}_{24} + \Omega^2 \hat{H}_{44} \right], \quad (97) \]

In this case, when the mechanical loading is in mode I type only, Eq. (93) is reduced to
\[ J_C^l = \frac{1}{4} \left( \frac{K_{\sigma,C}^a}{H_{22}} \right)^2 + \frac{1}{2} \left[ H_{22} \Lambda_3 (\Omega - 1) + H_{24} \Omega \right] K_{\sigma,C}^a K_{E,C}^a / 2 + \Theta(K_{E,C}^a)^2 = \Gamma. \] (98)

On the other hand, we may rewrite Eq. (95a) as

\[ K_{E,C}^a = \pm \sqrt{\frac{\Gamma}{\Theta}}, \] (99)

where the positive and negative signs are used for positive and negative electrical fields, respectively. Then, using Eqs. (94), (97) and (99), we rewrite Eq. (98) in a dimensionless form:

\[ \left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^a} \right)^2 + \left( \frac{H_{22} \Lambda_3 (1 - \Omega) - H_{24} \Omega}{\sqrt{H_{22} \Theta}} \right) \left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^a} \right) \left( \frac{K_{E,C}^a}{K_{E,C}^a} \right) + \left( \frac{K_{E,C}^a}{K_{E,C}^a} \right)^2 = 1. \] (100)

Equation (100) is the failure criterion derived from the general piezoelectric formulation for conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading. Comparing Eq. (100) with Eq. (40) indicates that both equations have the same form, representing an elliptic curve in the dimensionless coordinator system, as described after Eq. (40). In Part II of the series, Equation (100) will be used to compare with experimental results on the failure behavior of conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading.

3.4. Concluding remarks
Chapter 3 Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric Ceramics: Charge Free Zone (CFZ) Model

The CFZ model is extended to predict the failure behavior of conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading. In the CFZ model, piezoelectric ceramics are treated mechanically brittle and electrically ductile such that charge emission and charge trapping are assumed to occur at the conductive crack tip. The trapped charges partially shield the crack tip from applied electrical field and the local electric intensity factor has a non-zero value. Consequently, a non-zero local electric energy release rate contributes to the driving force to propagate the conductive crack. The merit of the CFZ model, similar to the DFZ model, lies in the ability to apply the Griffith criterion directly to link the local energy release rate to the fracture toughness in a completely brittle manner. The CFZ model yields an explicit failure criterion to predict the failure behavior of conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading. Mathematically, the failure formula takes form of $x^2 + \eta xy + y^2 = 1$ with $x = K_{\sigma,c}^a / K_{\sigma,c}^o$ (or $x = K_{\sigma,c}^a / K_{E,C}^o$) and $y = K_{\sigma,c}^a / K_{E,C}^o$ (or $y = K_{\sigma,c}^a / K_{E,C}^o$). The mathematic equation can be expressed in the standard form of an ellipse, $\hat{x}^2 /[2/(2+\eta)] + \hat{y}^2 /[2/(2-\eta)] = 1$, where the $(\hat{x}, \hat{y})$ coordinate system is established by rotating $45^\circ$ from the horizontal axis of the $(x, y)$ coordinate system.

If applied electric fields are parallel to the poling direction, $\eta < 0$ and the major semi-axis is located on the $\hat{x}$-axis, while $\eta > 0$ and the minor semi-axis is located on the $\hat{x}$-axis when applied electric fields are anti-parallel to the poling direction. For dielectric materials, $\eta = 0$ and the failure criterion is reduced to the failure criterion for conductive cracks in dielectric materials [16].
Chapter 3 Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric Ceramics: Charge Free Zone (CFZ) Model

References


Chapter 3 Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric Ceramics: Charge Free Zone (CFZ) Model


[27] Mihara T and Watabe H. Electronic Conduction Characteristics of Sol-Gel Ferroelectric Pb(Zr0.4Ti0.6)O3 Thin-Film Capacitors: Part I, Jpn J Appl Phys 1995;34:5664.


Fig. 3.1. A single line charge in front of an electrically conductive crack subjected to the image force and the tip driving force.
Fig. 3.2. The field distribution in front of a conductive crack, wherein $ob$ is the size of the charge-free zone and $ba$ denotes the charge zone.
\[ J^M = C_M \sigma^2 a \]
\[ J^E = C_E E^2 a \]

Fig. 3.3 A comparison of a normal crack loaded by uniform mechanical stress, \( \sigma \), and a conductive crack loaded by uniform electrical field, \( E \), where \( J^M \) and \( J^E \) denote, respectively, the mechanical and electrical \( J \)-integrals, and \( C_M = \pi / y \) and \( C_E = \pi \kappa / 2 \).
Chapter Four

Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric ceramics:
Experimental Verification

4.1. Introduction

Piezoelectric ceramics are widely used in smart structures like actuators and sensors et al. The embedded electrodes in smart structures may act as the conductive cracks and cause the failures of smart structures. The failures of smart structures may be treated as conductive crack problems. Therefore, it is of great interests to study the conductive crack problems of piezoelectric ceramics. Since there are many similarities between dielectric breakdown and fracture, many researchers have put great efforts trying to apply the concepts of fracture mechanics to the failure of dielectric and piezoelectric materials under mechanical and/or electrical loading [1-9]. Currently, the critical electrical field strength, called the dielectric breakdown strength, is used to serve as a failure criterion to predict dielectric breakdown. The dielectric breakdown strength is similar to the ultimate fracture strength, which is very sensitive to defects such as voids, flaws, and microcracks. These defects, however, exist in most materials and thus make the failure criterion of the ultimate fracture strength inappropriate. To provide failure criteria of materials with cracks, fracture mechanics has been quickly developed and found wide applications in industrial and engineering practice. Regarding dielectric failures, experimental
results have revealed that mechanical stresses enhance the failure process of
dielectric breakdown [10, 11] and voids play a significant role in the degradation of
dielectric breakdown [12], thereby indicating that the characteristics of the dielectric
breakdown strength are similar to the characteristics of the mechanical fracture
strength. Furthermore, electric field at a conductive crack tip theoretically
approaches infinity [5, 13], which is similar to the stress singularity at a conventional
 crack tip. All these similarities between dielectric breakdown and fracture suggest
that fracture mechanics should be able to treat successfully failures of dielectric and
piezoelectric solids under mechanical and/or electrical loading.

In the endeavor of applying fracture mechanics to failures of dielectric and
piezoelectric solids under mechanical and/or electrical loading, well controlled and
designed experiments play a crucial role. The experimental results on conductive
 cracks in depoled and poled PZT ceramics under purely electrical loading have
evidenced the existence of the electric fracture toughness [6, 7]. The comprehensive
investigation of the failure behavior of conductive cracks in thermally depoled PZT
 ceramics under mechanical and/or electrical loading indicates that the failure
behavior follows a circular curve in terms of the normalized critical stress and
electric intensity factors, which is predicted by the charge-free zone (CFZ) model
[8]. The CFZ model is based on the field limiting space charge (FLSC) model [14]
and in analogy with the dislocation-free zone (DFZ) model [15-17] and the tip-
emission-adjusted zone (TEAZ) model [18, 19] in the plastic fracture mechanics.
Various techniques, which include the thermal pulse, laser-intensity modulation,
laser induced pressure pulse, pressure wave propagation, non-structures acoustic
pulse, acoustic probe and piezoelectrically generated pressures steps, have been
applied to study the charge carrier types and the behavior of the space charge trapped in dielectrics. The detailed measurement methods can be found in a review paper [26]. Recently, a novel technique, electrostatic force microscopy (EFM), which is a modified atomic force microscopy (AFM) is implemented to study the space charge carrier behavior in dielectrics like silicon dioxide and semi-conductor materials like poly-silicon and gallium nitride [27-33]. Observations revealed that injected charge carriers can be trapped by the grain boundaries [27], interfacial surfaces [28] where huge crystal flaws exist. Dislocations can also act as the carrier trapping sites in gallium nitride [29, 31].

The CFZ model was extended to piezoelectric ceramics [20]. The simplified and general constitutive relationships were adopted in the theoretical analysis in order to illustrate clearly the physical picture [13, 20, 21]. With the general constitutive equation, the CFZ model yields the failure criterion [20]

\[
\left( \frac{K_{\sigma, C}^a}{K_{\sigma, C}^o} \right)^2 + \frac{\hat{H}_{22} \Lambda_2 (1 - \Omega) - \hat{H}_{24} \Omega}{\sqrt{\hat{H}_{22} \Theta}} \left( \frac{K_{\sigma, C}^a}{K_{\sigma, C}^o} \right) \left( \frac{K_{E, C}^a}{K_{E, C}^o} \right) + \left( \frac{K_{E, C}^a}{K_{E, C}^o} \right)^2 = 1, \tag{1}
\]

where \( K_{\sigma} \) denotes \( K_1, \hat{H}_{22}, \hat{H}_{24}, \Lambda_2 \), and \( \Theta \) are constants related to the material constants and the sample orientation and the parameter, \( \Omega \), which are described in detail in Part I, \( K_{\sigma, C}^o \) and \( K_{E, C}^o \) denote the mechanical fracture toughness and electrical fracture toughness under purely mechanical and electrical loading, respectively, and \( K_{\sigma, C}^a \) and \( K_{E, C}^a \) stand for the critical values of the applied stress and
electrical intensity factors at failure under combined mechanical and electrical loading, respectively. The parameter, $\Omega$, is defined as

$$
\Omega = \sqrt{\frac{b}{a}} \frac{F\left(\frac{\pi}{2}, \sqrt{1 - \frac{b}{a}}\right)}{E\left(\frac{\pi}{2}, \sqrt{1 - \frac{b}{a}}\right)},
$$

(2)

where $ob$ and $ba$ denote the sizes of the charge-free zone and the charge zone, respectively, as shown in Fig.4.2 in Chapter 3, and $F(\frac{\pi}{2},k)$ and $E(\frac{\pi}{2},k)$ are the complete elliptic integral of the first and second kinds, respectively with $k = \sqrt{1 - \frac{b}{a}}$. In Eq. (1), the negative sign is for positive electric loading and positive sign is for negative electric loading. Mathematically, Eq. (1) takes the form of $x^2 + \eta xy + y^2 = 1$ with $x = K_{\sigma,c}^a / K_{\sigma,c}^e$ (or $x = K_{\varepsilon,C}^a / K_{\varepsilon,C}^e$) and $y = K_{\varepsilon,C}^a / K_{\varepsilon,C}^e$ (or $y = K_{\sigma,C}^a / K_{\sigma,C}^e$), and $\eta$ being a constant. The mathematic equation can be expressed in the standard form of an ellipse, $\hat{x}^2 / [2/(2 + \eta)] + \hat{y}^2 / [2/(2 - \eta)] = 1$, where the $(\hat{x}, \hat{y})$ coordinate system is established by rotating 45° from the horizontal axis of the $(x, y)$ coordinate system, which describes an ellipse in terms of the normalized applied intensity factors. If the poling direction is along the $x$-direction, which is used in the modeling in Part I of this series, $\eta < 0$ and the major semi-axis is located on the $\hat{x}$-axis under positive electrical loading, while $\eta > 0$ and the minor semi-axis should be located on the $\hat{x}$-axis under negative electrical loading. For dielectric materials, the piezoelectric constants are zero and thus, the interaction term, i.e., the second term on the left hand-side of Eq. (1) disappears,
thereby reducing Eq. (1) to the failure criterion for conductive cracks in dielectric materials [8]. In the present work, we report experimental results on conductive cracks (deep notches) in PZT-8 piezoelectric ceramics under electrical and/or mechanical loading. The experimental results verify the theoretical predictions.

4. 2. Experiment

4. 2. 1. Sample preparation and fracture tests under electrical and/or mechanical loading

Piezoelectric ceramics of PZT series have great application in smart systems due to their higher piezoelectric constants. The material used in this study was poled lead zirconate titanate ceramics (PZT-8, Morgan ElectroCeramics). Compared to PZT 4, piezoelectric ceramics PZT 8 has lower mechanical losses under high electric drive. Since it is difficult to make pre-cracks in ceramic samples, we used pre-notched compact tension (CT) samples in the fracture tests under purely mechanical, purely electrical and mixed mechanical and electrical loads. Figure 4.1 schematically shows the sample geometry, the poling direction, and the loading conditions. All samples had widths of \( w = 10.0 \) mm, heights of \( h = 10.0 \) mm and thicknesses around \( d = 2.5 \) mm. A pre-notch or crack was cut in each sample with a 0.3 mm thick diamond saw and further sharpened by a 0.12 mm thick diameter saw. The total notch length varied from sample to sample and ranged from 4.58 mm to 6.35 mm, which corresponded to the ligament, \( l \), ranging from 5.43 to 3.65 mm. Comparing the notch length with the notch radius reveals that the notches are all deep and sharp. Fracture mechanics [22] has shown that, for a deep and sharp notch, the stress field near the notch tip can also
be expressed in terms of the stress intensity factor and the relationship between the mechanical energy release rate and the stress intensity factor holds. We believe that the failure behavior of the conductive deep and sharp notches can represent the failure behavior of conductive cracks except that the values of the critical stress intensity factor and the critical electric intensity factor may vary in some extent. After the cutting, the samples were cleaned ultrasonically in de-ionized water for ten minutes. To create conductive cracks, silver paint was filled into the notch (crack) to make it function as an electrode. The silver paint is a viscous emulsion and solidifies at room temperature quickly. A refilling technique, which meant that filling was conducted again after the preceding filling and solidification, was adopted to ensure that the notch was fully filled. The refilling continued until no obvious shrinkage was observed by the naked eye. Copper wires were connected to the upper and bottom surfaces using a kind of conductive adhesive (RS Company), which was actually silver-added epoxy. Then, the samples were put into an oven at 55 °C for 24 hours to solidify and stabilize the adhesive.

4. 2.2. Finite element calculations

Finite element calculations were conducted to establish the mode I stress intensity factor, $K_I$, and the electric intensity factor, $K_E$, for the used sample geometry and loading conditions. We used the commercial software ANSYS and the plane strain piezoelectric elements (Plane 13 in ANSYS). The linear constitutive relationship, i.e. Eq. (41) in Chapter two, was adopted. The material constants used in the numerical
calculations for the PZT-8 are provided by the manufacturing company and listed below:

<table>
<thead>
<tr>
<th>Stiffness Constants ( (10^{10}\text{ Nm}^{-2}) )</th>
<th>Piezoelectric Constants ( (C\text{m}^{-2}) )</th>
<th>Relative Dielectric Constants ( (\kappa / \kappa_0) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( c_{11} = 14.9 ), ( c_{33} = 13.2 )</td>
<td>( e_{31} = -4.1 )</td>
<td>( \kappa_{11} = \kappa_{22} = 1290 )</td>
</tr>
<tr>
<td>( c_{44} = 3.13 )</td>
<td>( e_{33} = 13.2 )</td>
<td>( \kappa_{33} = 1000 )</td>
</tr>
<tr>
<td>( c_{12} = c_{13} = 8.11 )</td>
<td>( e_{15} = 10.3 )</td>
<td>( \kappa_{0} = 8.855 \times 10^{-12} \text{Cm}^2 / \text{V} )</td>
</tr>
</tbody>
</table>

The direct method was used to calculate stress and electric intensity factors for the CT samples under mechanical and/or electrical loading, which required very fine mesh near the crack tip in order to obtain numerical results with high accuracy. The boundary conditions were applied based on the actual experimentation. Point forces \( F \) and voltage \( V \) were applied as shown in Fig 4.2 a. Conventional (nonsingular) elements were used to model the crack-tip singularity. Figure 4.2 shows the mesh used in the finite element analysis, in which over 13,000 elements were adopted. The mesh was step by step refined by five steps with each step reducing the element size by one order in magnitude. The side length of the elements near the crack tip was \( 2.0 \times 10^{-5} \text{ mm} \), which was roughly at the level of one millionth of the sample length. After having established the stress field \( \sigma_y \), electric field \( E_x \) in the proximity of the crack tip, the stress intensity factor can be numerically estimated. Both \( \sigma_y \) and \( E_x \) are local values. For mode I crack, stress intensity factor, \( K_\sigma \), and the electric intensity factor, \( K_E \), were evaluated from

\[
K_\sigma = \sqrt{2\pi x} \sigma_y(x, y = 0), \quad 0 < x < \delta, \quad (3a)
\]
where $x$ is the distance from the crack tip in the $x$-direction and $\delta$ is a sufficiently small length. The values of $\sqrt{2\pi x} \sigma_y(x, y = 0)$ and $\sqrt{2\pi x} E_x(x, y = 0)$ remain almost constant in a large region of the vicinity at the crack front, as shown in Fig.4. 3, except a few points close to the crack tip. This is because that the stress singularity and the electric field singularity make the numerical results unreliable on the few points close to the crack tip. The constant values of $\sqrt{2\pi x} \sigma_y(x, y = 0)$ and $\sqrt{2\pi x} E_x(x, y = 0)$ were taken as stress and electric intensity factors, respectively. Finally, the numerical results were fitted into the following formulas of the intensity factors for the nominal crack length, $c=w-l$, ranging from 2.75 to 6.50 mm:

\[ K_\sigma = \frac{P}{d\sqrt{w}} f_1 \left( \frac{c}{w} \right) + k_{11} \left( \frac{c}{w} \right) e_{33} \frac{V}{\sqrt{w}} , \quad (4a) \]

\[ K_E = \frac{V}{\sqrt{w}} f_2 \left( \frac{c}{w} \right) + k_{22} \left( \frac{c}{w} \right) \frac{P}{d\sqrt{w}} e_{33} , \quad (4b) \]

with

\[ f_1 \left( \frac{c}{w} \right) = -37.96 + 281.23 \left( \frac{c}{w} \right) - 634.11 \left( \frac{c}{w} \right)^2 + 590.85 \left( \frac{c}{w} \right)^3 - 123.25 \left( \frac{c}{w} \right)^4 , \quad (4c) \]

\[ f_2 \left( \frac{c}{w} \right) = -2.284 + 15.388 \left( \frac{c}{w} \right) - 22.693 \left( \frac{c}{w} \right)^2 + 10.152 \left( \frac{c}{w} \right)^3 + 4.545 \left( \frac{c}{w} \right)^4 , \quad (4d) \]
\[ k_{11} = 3.418 + 30.154\left(\frac{c}{w}\right) + 96.230\left(\frac{c}{w}\right)^2 - 132.153\left(\frac{c}{w}\right)^3 + 64.635\left(\frac{c}{w}\right)^4, \]

\( (4e) \)

\[ k_{22} = 4.194 - 49.304\left(\frac{c}{w}\right) + 208.269\left(\frac{c}{w}\right)^2 - 345.077\left(\frac{c}{w}\right)^3 + 206.016\left(\frac{c}{w}\right)^4, \]

\( (4f) \)

where \( w = 10^{-2} \text{ m} \), \( P \) is the mechanical load in units of Newton, \( d \) is the thickness in units of meter, and \( V \) is the applied voltage in units of Voltage and the dimensionless parameter, \( c/w \), ranges from 0.275 to 0.650.

In the used CT sample, there is a slot designed for mechanical loading, which may arise the question how to define the crack or notch length. In the present study, we define the nominal notch or crack length as \( c = w - l \), as shown in Fig. 4.1, which includes the size of the mechanical loading slot along the notch direction. Since the mechanical loading slot has a considerable size, the mechanical loading slot might influence the calculated values of intensity factors, \( K_\sigma \) and \( K_E \). To study this effect, we calculated \( K_\sigma \) and \( K_E \) for an ideal CT sample without the mechanical loading slot, as shown in Fig. 4.2(b). The calculation results indicate that with the same loading conditions and the value of \( w - l = 3.5 \text{ mm} \), the calculated stress intensity factor, \( K_\sigma \), has the same value of 172.0 \( Pa\sqrt{m} \) for both the used and ideal CT samples, whereas the calculated value of the electric intensity factor is 17.3 \( V/\sqrt{m} \) for the used CT sample, which is slightly lower than that of 17.4 \( V/\sqrt{m} \) for the ideal
CT sample. The results indicate that the mechanical loading slot may slightly lower
the electric intensity factor in the used CT samples in comparison with the ideal CT
sample and the influence on the stress intensity factor is negligible.

4. 2.3. Numerical calculations of the parameters used in the CFZ model

Using the materials constants for the PZT-8 ceramics and letting the poling
direction parallel to the negative $x$-direction, the constitutive equations take the form
of

\[
\begin{bmatrix}
\sigma_{11} \\
\sigma_{22} \\
\sigma_{33} \\
\sigma_{23} \\
\sigma_{31} \\
\sigma_{12}
\end{bmatrix} = \begin{bmatrix}
c_{33} & c_{13} & c_{13} & 0 & 0 & 0 \\
c_{13} & c_{11} & c_{12} & 0 & 0 & 0 \\
c_{13} & c_{12} & c_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{c_{11} - c_{12}}{2} & 0 & 0 \\
0 & 0 & 0 & 0 & c_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & c_{44}
\end{bmatrix}
\begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{23} \\
2\varepsilon_{31} \\
2\varepsilon_{12}
\end{bmatrix} = \begin{bmatrix}
-e_{33} & 0 & 0 \\
-e_{13} & 0 & 0 \\
-e_{13} & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & -e_{15} \\
0 & 0 & 0
\end{bmatrix}
\begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix}
\]

(5a)

\[
\begin{bmatrix}
D_1 \\
D_2 \\
D_3
\end{bmatrix} = \begin{bmatrix}
-e_{33} & -e_{13} & -e_{13} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -e_{15} \\
0 & 0 & 0 & 0 & -e_{15} & 0
\end{bmatrix}
\begin{bmatrix}
\varepsilon_{11} \\
\varepsilon_{22} \\
\varepsilon_{33} \\
2\varepsilon_{23} \\
2\varepsilon_{31} \\
2\varepsilon_{12}
\end{bmatrix} + \begin{bmatrix}
\kappa_{33} & 0 & 0 \\
0 & \kappa_{11} & 0 \\
0 & 0 & \kappa_{22}
\end{bmatrix}
\begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix}
\]

(5b)

If the poling direction is along the positive $x$-direction, the minus signs of the
piezoelectric constants should be taken away. To have a smooth mathematical
 calculation of the eigen-equation, i.e., Eq. (47) in Part I of this series, we took new

62
units or scales marked with (*) by letting $C_{\alpha\beta}^* = 10^{-9} C_{\alpha\beta}, \quad \varepsilon_{\alpha}^* = 10^9 \varepsilon_{\alpha}, \quad D_i^* = 10^9 D_i, \quad \kappa_{\alpha}^* = 10^9 \kappa_{\alpha}$ and other properties and constants having the same units or scales as the original ones. In this case, we solved the eigen-equation and chose the eigen-vector matrices, $\mathbf{A}$ and $\mathbf{B}$, satisfying Eq. (48) in Part I of this series. Then, we constructed matrices $\hat{\mathbf{A}}$ and $\hat{\mathbf{B}}$ from matrices $\mathbf{A}$ and $\mathbf{B}$ and calculated matrices $\hat{\mathbf{Y}}$ and $\hat{\mathbf{H}}$. Matrix $\hat{\mathbf{H}}$ is given by

$$
\hat{\mathbf{H}} = \begin{pmatrix}
0.0416 & 0 & 0 & 0 \\
0 & 0.0394 & 0 & -0.2760 \\
0 & 0 & 0.0613 & 0 \\
0 & -0.2760 & 0 & 26.30
\end{pmatrix}.
$$

(6)

The effective dielectric constant and the vector, $\Lambda$, were also calculated to be

$$
\kappa_{\alpha} = \frac{i}{\Lambda_4 \Lambda_4^* - \Lambda_4^* \Lambda_4} = 12.186,
$$

(7a)

$$
(\Lambda_1 \quad \Lambda_2 \quad \Lambda_3 \quad \Lambda_4) = (0 \quad -7.001 \quad 0 \quad 1).
$$

(7b)

Then, the parameter, $\Theta$, takes the explicit form:

$$
\Theta = \frac{1}{4} \left[ \Lambda_1^2 (\Omega + 1)^2 \hat{H}_{22} + 2 \Lambda_1 (\Omega + 1) \Omega \hat{H}_{24} + \Omega^2 \hat{H}_{44} \right] \approx 8.0239 \Omega^2 - 1.9317 \Omega + 0.4828.
$$

(8)

Finally, Eq. (1) can be explicitly expressed as
\[
\left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^o} \right)^2 \mp \frac{0.5518\Omega - 0.2758}{\sqrt{0.3161\Omega^2 - 0.0761\Omega + 0.0190}} \left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^o} \right) \left( \frac{K_{E,C}^a}{K_{E,C}^o} \right) + \left( \frac{K_{E,C}^a}{K_{E,C}^o} \right)^2 = 1.
\]

(9a)

Equation (9a) is going to compare with the experimental results. It should be pointed out that if the poling direction is along the positive \(x\)-direction, the components of matrix \(\hat{H}\) have all the same values except that \(\hat{H}_{24}\) and \(\hat{H}_{42}\) change the sign from minus to plus. In this case, the effective dielectric constant remains unchanged and the parameter, \(\Lambda_2\), has the same absolute value but change the sign from minus to plus. As a result, the \(\Theta\), takes the explicit form of Eq. (8), but the failure criterion, Eq. (9a), changes to

\[
\left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^o} \right)^2 \mp \frac{0.2758 - 0.5518\Omega}{\sqrt{0.3161\Omega^2 - 0.0761\Omega + 0.0190}} \left( \frac{K_{\sigma,C}^a}{K_{\sigma,C}^o} \right) \left( \frac{K_{E,C}^a}{K_{E,C}^o} \right) + \left( \frac{K_{E,C}^a}{K_{E,C}^o} \right)^2 = 1.
\]

(9b)

4.2.4. Fracture tests under electrical and/or mechanical loading

The fracture tests were carried out under a constant voltage mode or a constant mechanical load mode with a homemade loading apparatus (see Fig.4.10). In the constant voltage mode, a constant voltage was applied first; then, the mechanical
load was gradually increased by increasing the weight until the sample fractured. In the constant mechanical load mode, a constant mechanical load was applied first, followed by a gradual increase of the applied electric voltage until the sample fractured. To avoid electric sparking, a thick layer of silicone grease was put on the sample surfaces. In the present study, all tests were conducted at room temperature. The critical load and the critical voltage at fracture were recorded to calculate the critical stress intensity factor and the critical electric intensity factor. Sparkling lights were observed in the dielectric breakdown tests as shown in Fig. 4.11.

4.3. Experimental results and discussion

Figure 4. 4(a) shows the relationship of the critical load versus the crack length for the experimental results under purely mechanical loading, where a solid circle represents an experimental datum and hereafter the same symbol is used in the rest figures without notation. Using Eq. (4a) and the critical loads at fracture, we calculate the critical stress intensity factors and plot them, as a function of the crack length, in Fig. 4.4(b). The mean and standard deviation of the critical stress intensity factor is \( K_{\sigma,C}^0 = 1.26 \pm 0.06 \text{ MPa}\sqrt{m} \) under purely mechanical loading. The value of \( K_{\sigma,C}^0 = 1.26 \text{ MPa}\sqrt{m} \) is higher than the fracture toughness, \( K_{\sigma,C}^0 = 0.934 \text{ MPa}\sqrt{m} \), of the poled PZT-4 ceramics [23], but within the range, \( K_{\sigma,C}^0 = 1.1 - 1.7 \text{ MPa}\sqrt{m} \), reported by Freiman and White [24]. From the mean and the standard deviation, we have the relative error, \( \pm 4.8\% \), in the measured mechanical fracture toughness.
An applied electric field could fracture the poled ceramic samples with conductive deep and sharp notches. The fracture was accompanied with dielectric discharging. Similar to the depoled PZT ceramics, when the deep notch of a sample was free of silver paint, the sample could survive an electric voltage of 20 kV, which was the maximum voltage that the used power supplier could provide. After filling up the notch with silver paint, the notch became electrically conductive. Then, an applied electric field about 11.4 kV fractured the sample. For samples of short ligaments, cracks always propagated along the notch direction. For some samples of long ligaments, the crack propagate was not so straightforward, as shown in Fig.4. 5 (b), where the straightforward crack propagation under purely mechanical loading was shown in Fig.4. 5(a) for comparison. The straightforward crack propagation mode implies a flat fracture surface, while a curved crack propagation mode indicates a rough fracture surface, which will be discussed later. As described in the previous works [6, 8], the charges in the upper and lower surfaces of a conductive crack have the same sign. The charges with the same sign repel each other and generate a Coulombic force that opens and propagates the conductive crack. For the CT samples of the brittle ceramics, the fracture process is unstable because the stress and electric intensity factors increase both monotonically with the crack length, which means that failure will occur once the crack propagation is triggered mechanically and/or electrically. The fracture process and the dielectric breakdown may occur simultaneously in the CT samples, even so charge emission from the conductive notch (crack) tip may happen prior to the process of fracture and dielectric breakdown. It is the sample geometry and the loading condition that makes the failure become two-dimensional. The experimental results all reveal the two-dimensional failure behavior, as shown in Fig.4.5. In the present work, we study only
the onset of the failure process and the critical values of the voltage and the mechanical load at the onset are used to calculate the electrical fracture toughness and the mechanical fracture toughness.

As mentioned above and shown in Fig. 4.1, the poling direction is along the negative x-direction for the used samples. The experimental results under purely negative electrical loading are plotted in Fig. 4.6(a), showing the relationship of the critical voltage at fracture versus the deep notch length. Using Eq. (4b) and the critical voltages, we calculate the critical electric intensity factors and plot them, as a function of the deep notch length, in Fig. 4.6(b). The mean and standard deviation of electric fracture toughness is \( K_{e,C}^0 = 165.2 \pm 12.7 \text{ kV} / \sqrt{m} \) for the poled ceramics under purely negative electrical loading. Similarly, Fig.4.7(a) shows the relationship of the critical voltage at fracture versus the deep notch length under purely positively electrical loading and Fig. 4.7(b) illustrates the critical electric intensity factor as a function of the deep notch length. From the experimental data, we have the mean and standard deviation of electric fracture toughness to be \( K_{e,C}^0 = 157.1 \pm 14.1 \text{ kV} / \sqrt{m} \) for the poled ceramics under purely positive electrical loading. The difference between the means of the electrical fracture toughness under purely positive and negative electrical loading is small in comparison with the standard deviations. This result implies that the major driving force to propagate the conductive cracks is the statically electric force between the upper and lower surfaces of the cracks and the piezoelectric effect does not play a substantial role in the failure under purely electric loading. However, the piezoelectric effect does affect the failure behavior of conductive cracks under combined electrical and mechanical loading, which will be
described later. Like the mechanical fracture toughness, the electric fracture toughness is a material constant independent of the crack length and thus can serve as a failure criterion for conductive cracks (or deep notches) in piezoelectric ceramics under purely electrical loading. The relative errors in the experimental data of the electric fracture toughness under purely positive and negative electrical loading are about ±9.0% and ±7.7%, respectively, which are higher than that of the mechanical fracture toughness of about ±4.8%. The experimental errors may be attributed to the following reasons: 1) the notch is not sharp enough, 2) there are many defects such as grain boundaries and pores in the PZT ceramics fabricated by sintering PZT powders, and 3) there may exist electric defects in the PZT ceramics, which cause partially electric discharging, etc.

To compare the electrical fracture toughness with the mechanical fracture toughness, we convert them correspondingly to the critical values of the energy release rates by using Eq. (67) in Part I of this series. The mean values of the critical energy release rate under purely positive and negative electric loading are $G_{e^+,C} = 164.3 \, N/m$ and $G_{e^-,C} = 181.6 \, N/m$, whereas the mean value of the critical energy release rate is $G_{e,C} = 15.9 \, N/m$ under purely mechanical loading. The electrical toughness under either purely positive or negative electric loading is over ten times higher in magnitude than the mechanical toughness. The high electrical toughness is attributed to electrical plastic deformation, such as electrical discharge, etc, that occurs at the tip of the conductive notch, forming an electrical plastic zone. Actually, dielectric breakdown accompanies the fracture under purely electrical loading. The fracture surfaces are flat for samples fractured under purely mechanical loading, while purely
electrical loading yields rough fracture surfaces. In the electrically fractured samples, the discharge may locally burn the sample. It is the electrical plastic deformation that consumes more energy and thus leads to the high electrical fracture toughness. Furthermore, the mechanical toughness may be regarded as the critical value of the local energy release rate, $J^c$, as indicated by Eq. (94) in Part I of this series. If we use the local energy release rate as a failure criterion, as we do in the theoretical modeling, the experimental result that the electric toughness is much higher than the mechanical toughness gives a hint that a shielding mechanism must take place under electrical loading.

From the experimental results under purely electrical or mechanical loading, we estimate the value of the parameter, $\Omega$, from Eqs. (96) and (97) in Part I of this series, to be 0.303 under purely positive electrical loading and 0.281 under purely negative electrical loading. Then, using Eq. (2), we estimate the value of the ratio, $b/a$, which is 0.0048 under purely positive electrical loading and 0.0039 under purely negative electrical loading, while the $b/a$ value is 0.0028 for the thermally depoed PZT-4 ceramics.

The critical loads and the critical voltages at fracture under combined mechanical and electrical loading are tabulated in Tables 1 and 2 for the constant voltage and constant mechanical load modes, respectively. As indicated in Tables 1 and 2, the data become more scattering than those under purely mechanical or electrical loading. For instance, for the 9 samples with the almost same notch length of 4.64 ± 0.05 mm, the value of the mechanical load varies from 21.8 N to 29.3 N under a constant voltage of 2.0 kV. The phenomenon that combined mechanical and
electrical loading leads to an increase in the experimental error has been observed before in the bending tests on PZT-841 ceramics [13, 25] and in the fracture tests on depoed PZT-4 ceramics [8]. In the bending tests on the PZT-841 ceramics, the mechanical bending strength at an applied electric field of 10 kV/cm varies from about 30 MPa to about 120 MPa with a relative error of about 33%. The value of the mechanical load varies from 18.4 N to 29.0 N for the 17 samples with the almost same notch length of 5.04 ± 0.19 mm in the fracture tests on the depoed PZT-4 ceramics under a constant voltage of 3.0 kV [8]. Due to the large scattering of the experimental data, we tested sufficiently large number of samples to ensure statistical assessment of the failure behavior. From the experimental data and the crack length, we calculate the critical stress intensity factor, $K_{\sigma,C}^a$, and the critical electric intensity factor, $K_{E,C}^a$, for each sample. Then, we normalize the critical stress intensity factor by the critical stress intensity factor under purely mechanical loading, and normalize the critical electric intensity factor by the critical electric intensity factor under purely electric loading. Figures 4.8(a) and 4.8(b) show the relationships of the normalized electric intensity factor versus the normalized stress intensity factor under combined mechanical and electrical loading with (a) for negative and (b) for positive electric fields. As described above, the CFZ model predicts that the failure behavior should have a shape of an ellipse. We adopt the least square method to fit the experimental data with the mathematic equation, $x^2 + \eta xy + y^2 = 1$. The fitting results are illustrated by the dashed curves in Figs. 4.8(a) and 4.8(b), where unit circles are plotted by solid curves for comparison. The fitting results indicate that $\eta = -0.16$ and the major semi-axis of the ellipse is on the line of $K_{\sigma,C}^a / K_{\sigma,C}^0 = K_{E,C}^a / K_{E,C}^0$ under negative electrical loading, and $\eta = 0.52$ and the
minor semi-axis of the ellipse is on the line of \( K^{a}_{\sigma,C} / K^{0}_{\sigma,C} = K^{a}_{E^*,C} / K^{0}_{E^*,C} \) under positive electrical loading, which qualitatively verify the predictions of the CFZ model for piezoelectric ceramics. Using the estimated value of \( \Omega \), we calculate the theoretical value of \( \eta \) from \( \eta = \frac{0.5518 \Omega - 0.2758}{\sqrt{0.3161 \Omega^2 - 0.0761 \Omega + 0.0190}} \) and the results are \( \eta = 0.69 \) under positive electrical loading and \( \eta = -0.80 \) under negative electrical loading. Comparing the theoretical predictions with the corresponding experimental results indicates that the experimental data could be approximately described by Eq. (1). The difference between the theoretical predictions and the experimental results might be attributed to the simplification of the complicated failure behavior in the CFZ model and to the large scattering in the measured data. Nevertheless, the theoretical and experimental results indicate that Eq. (1) is able to serve as a failure criterion for the poled PZT ceramics with conductive cracks under electrical and/or mechanical loading.

Figure 4.9 shows fractographs for the samples fractured under different loading conditions. Under purely mechanical loading, the fracture surface was flat, as shown in Fig. 3.9(a). Under purely electrical loading, the fracture surface was very rough and most of the fracture surface was melted and re-solidified, as evidenced by the dark area in Fig. 3.9(d). As expected, the morphology of the fracture surface under combined electrical and mechanical loading was between these morphologies under purely mechanical loading and purely electrical loading. As shown in Figs. 4.9(b) and 4.9(c), the melted and re-solidified surface area increased as the level of the electrical loading increased. The melted and re-solidified surface region could take the shape of channels, as shown in Fig. 4.9(c) or occupied just a small area near the
notch front, as shown in Fig. 4.9 (b). The melted and re-solidified surface region was obviously caused by dielectric breakdown, at which large electric current pulses occurred and thus melted the ceramics. The difference in the shape of the melted and re-solidified surface region might be attributed to some defects, which were sensitive to dielectric breakdown.

4.4. Concluding remarks

The designed experiments of the failure behavior of electrically conductive cracks (deep notches) in poled lead zirconate titanate PZT-8 ceramics under electrical and/or mechanical loading verify the theoretical predictions from the charge-free zone model described in Part I of this series. The critical load and/or critical voltage at the onset of the failure were recorded to calculate the critical values of the stress and electric intensity factors and then the critical energy release rate. The critical energy release rates for the conductive cracks (deep notches) in the PZT-8 ceramic compact tension samples were 164.3 N/m and 181.6 N/m under purely positive and negative electrical fields, respectively, which were over ten times higher in magnitude than the critical energy release rate of the same samples under purely mechanical loading. The difference in the toughness is attributed to the electrical plastic deformation. The electrical plastic deformation might be charge emission and charge trapping at the crack tip. Thus, we may treat piezoelectric ceramics mechanically brittle and electrically ductile in the charge-free zone model. The explicit failure criteria derived from the CFZ model were verified by the experimental results. The failure criteria provide designers of electronic and
electromechanical devices with information on the electrical fracture toughness and the mechanical fracture toughness, thereby predicting the critical electric field and the critical mechanical load at which a piezoelectric ceramic material containing a conductive crack or an internal electrode fails under electrical and/or mechanical loading. The critical electric field and the critical mechanical load are functions of the crack dimensions or the length of the electrode, while the electrical fracture toughness and the mechanical fracture toughness are both material properties. Thus, one can predict the critical electric field and the critical mechanical load when information on the sample geometry and on the electrical and mechanical fracture toughness is available.
Chapter 4 Failure Behavior and Failure Criterion of Conductive Cracks (Deep Notches) in Piezoelectric ceramics: Experimental Verification

References

Table 4.1. Critical voltages with a pre-applied load, P, under positively electric loading

<table>
<thead>
<tr>
<th>Pre-applied P</th>
<th>Critical voltage (V)/Notch Length (mm)/Specimen Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P=0 N</td>
<td>9224/6.327/3.09, 13181/5.016/3.01, 9236/6.174/2.54, 9189/6.163/3.06, 12759/5.566/3.03, 11411/6.031/2.53, 9436/6.036/3.04, 10983/5.551/2.61, 12440/5.213/2.46, 11804/5.719/2.61, 12627/4.765/3.02, 8802/6.033/3.01, 8939/6.062/2.87, 9925/5.951/2.75, 11264/6.055/3.05, 8593/6.261/3.01, 9045/6.039/3.03, 8906/6.213/2.78, 8924/6.166/2.37, 11785/5.817/2.58, 10218/5.662/2.64, 9807/5.983/2.58, 12959/4.876/2.57</td>
</tr>
<tr>
<td>P=8.9 N</td>
<td>9493/4.715/2.46, 9539/4.540/2.85, 11345/4.679/2.54, 11009/5.591/2.31, 11502/5.349/2.55, 10724/5.342/2.48, 8644/4.624/2.81, 12700/4.720/2.31, 11075/4.602/2.29, 12612/4.649/2.62</td>
</tr>
<tr>
<td>P=11.9 N</td>
<td>11091/4.647/2.6, 10563/4.889/2.62, 12967/4.589/2.88, 13318/5.131/2.53</td>
</tr>
<tr>
<td>P=14.9 N</td>
<td>10662/4.612/2.66, 8661/4.672/2.82, 8661/5.321/2.87, 11370/4.597/2.56, 12751/6.326/2.59, 14966/4.647/2.88, 10693/4.617/2.48, 12288/4.582/2.47, 12164/4.703/2.38, 10991/5.313/2.74</td>
</tr>
<tr>
<td>P=20.2 N</td>
<td>10187/4.645/2.58, 7384/4.708/2.57, 9801/5.439/2.88, 12147/4.708/2.79, 8200/5.736/2.68, 8661/4.597/2.38, 8661/4.576/2.45, 10601/4.603/2.64, 12751/4.643/2.89, 1383//4.665/2.82</td>
</tr>
</tbody>
</table>
Table 4.2. Critical loads with a pre-applied voltage, $V$, under positively electric loading

<table>
<thead>
<tr>
<th>Pre-applied V</th>
<th>Critical Load (P)/Notch Length (mm)/Specimen Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>V=0</td>
<td>32.40/4.743/2.46, 30.16/5.457/2.53, 31.30/5.082/2.5, 35.12/4.596/2.97, 31.56/4.589/2.39, 33.94/4.790/2.62, 30.99/4.753/2.57, 34.41/4.747/2.56, 31.86/4.625/2.45, 33.13/4.604/2.52, 37.97/4.669/2.74, 34.47/4.665/2.59, 36.41/4.778/2.73, 36.37/4.780/2.68, 32.63/4.756/2.53, 35.13/4.710/2.65, 33.72/4.656/2.55, 32.84/4.766/2.51, 39.00/4.736/2.79, 37.86/4.701/2.77, 29.34/3.029/2.66, 26.57/3.031/2.61, 30.93/2.664/2.66, 29.27/2.832/2.65, 30.39/2.785/2.69, 27.14/3.015/2.61</td>
</tr>
<tr>
<td>V=1.5 kV</td>
<td>27.33/4.721/2.51, 26.42/4.593/2.61, 28.88/4.697/2.45, 28.21/4.637/2.61, 29.38/4.684/2.55, 31.18/4.731/2.66, 29.21/4.582/2.71, 24.39/4.702/2.34</td>
</tr>
<tr>
<td>V=2.0 kV</td>
<td>25.87/4.626/2.52, 21.76/4.661/1.96, 27.66/4.597/2.54, 27.84/4.616/2.43, 28.56/4.593/2.54, 24.67/4.585/2.40, 29.01/4.692/2.75, 29.31/4.706/2.57, 26.17/4.709/2.42</td>
</tr>
<tr>
<td>V=3.5 kV</td>
<td>25.97/4.539/2.80, 29.80/4.566/2.83, 28.88/4.577/2.69, 23.69/5.212/2.53, 21.47/5.158/2.76, 20.66/5.760/2.94, 22.88/4.756/2.56, 22.62/4.686/2.62</td>
</tr>
<tr>
<td>V=5.0 kV</td>
<td>21.98/5.916/2.52, 24.49/5.866/2.65, 17.57/5.961/2.57, 25.19/5.625/2.51, 22.49/5.832/2.85, 21.86/5.726/2.62, 12.15/6.098/2.64, 24.29/5.843/3.19</td>
</tr>
<tr>
<td>V=7.0 kV</td>
<td>17.18/5.425/2.94, 13.16/6.325/2.64, 20.26/5.839/2.68, 15.39/5.599/2.58, 16.16/6.099/2.67, 17.79/6.109/2.52, 12.27/6.052/2.59, 14.85/6.047/2.62, 16.18/6.060/3.02, 17.19/5.502/2.62</td>
</tr>
<tr>
<td>V=8.0 kV</td>
<td>8.62/6.027/2.97, 6.58/5.845/2.48, 7.34/6.036/2.48, 6.00/5.755/2.87, 4.13/5.889/2.55, 6.43/5.654/2.62, 6.15/5.995/2.59, 6.01/5.755/2.87, 3.48/5.983/2.61, 4.13/5.677/3.04</td>
</tr>
</tbody>
</table>
Table 4.3. Critical voltages with a pre-applied load, P, under negatively electric loading.

<table>
<thead>
<tr>
<th>Pre-applied P</th>
<th>Critical voltage (V)/Notch Length (mm)/Specimen Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P=0 N</td>
<td>10793/5.978/2.45, 12435/5.202/2.51, 11783/5.611/2.61, 11689/5.312/2.62, 11631/5.018/2.55, 11048/5.501/2.58, 13779/4.842/2.45, 14252/4.803/2.62, 11552/5.907/2.71, 9423/6.375/2.67, 12698/5.511/2.71, 11691/5.342/2.55, 11007/5.141/2.52, 10971/6.094/2.48, 11239/5.023/2.57, 13419/5.647/2.47, 10533/5.971/2.62, 12929/5.502/2.57, 11589/5.148/2.48, 11789/5.135/2.49, 11399/5.276/2.56, 14319/4.765/2.51, 13638/4.981/2.46, 11504/4.856/2.52, 11937/5.133/2.48, 10761/5.886/2.64</td>
</tr>
<tr>
<td>P=11.74 N</td>
<td>14574/5.016/2.56, 12541/4.802/2.56, 14155/4.869/2.58, 12470/5.186/2.46, 13738/5.331/2.63, 14097/4.717/2.37, 11882/4.737/2.46, 11283/4.792/2.41, 12525/4.828/2.59, 10874/4.815/2.65, 13552/4.729/2.64</td>
</tr>
<tr>
<td>P=15.38 N</td>
<td>12158/5.114/2.49, 12581/5.449/2.67, 12584/4.624/2.64, 11963/4.692/2.64, 11757/4.788/2.47, 12979/4.653/2.56, 13077/4.711/2.61, 12622/4.628/2.57, 10949/4.812/2.44</td>
</tr>
<tr>
<td>P=18.53 N</td>
<td>10930/4.853/2.57, 13180/4.748/2.47, 11173/4.719/2.46, 13133/5.449/2.56, 13404/4.789/2.53, 10458/5.649/2.55, 10692/4.753/2.59, 10506/4.856/2.47, 12322/4.789/2.49, 13390/4.713/2.48</td>
</tr>
<tr>
<td>P=24.46 N</td>
<td>11339/4.671/2.54, 10788/5.285/2.57, 10109/4.789/2.42, 8910/4.702/2.62, 10762/4.794/2.64, 10684/4.681/2.46, 10736/5.087/2.51, 13002/4.686/2.45, 7838/4.843/2.64, 8605/4.677/2.51</td>
</tr>
</tbody>
</table>
Table 4. Critical loads with a pre-applied voltage, V, under negatively electric loading

<table>
<thead>
<tr>
<th>Pre-applied V</th>
<th>Critical load (P)/Notch Length (mm)/Specimen Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>V=2.0 kV</td>
<td>25.88/4.896/2.39, 28.69/4.887/2.44, 29.47/5.351/2.61, 28.77/5.457/2.53</td>
</tr>
<tr>
<td></td>
<td>25.56/5.441/2.59, 28.34/5.307/2.57, 30.89/5.469/2.58</td>
</tr>
<tr>
<td>V=4.0 kV</td>
<td>25.03/5.252/2.51, 24.56/6.115/2.54, 23.56/5.371/2.56, 24.68/5.886/2.51, 26.63/5.322/2.51</td>
</tr>
<tr>
<td></td>
<td>27.87/5.455/2.53, 26.28/5.494/2.52, 23.47/5.421/2.49</td>
</tr>
<tr>
<td>V=6.0kV</td>
<td>20.69/6.274/2.52, 17.19/5.933/2.52, 15.39/6.247/2.41, 14.51/6.134/2.53, 18.79/5.934/2.46</td>
</tr>
<tr>
<td></td>
<td>23.33/5.779/2.51, 17.47/6.279/2.47, 17.61/5.779/2.53</td>
</tr>
<tr>
<td>V=8.0 kV</td>
<td>8.79/6.036/2.52, 9.15/6.241/2.42, 8.25/6.339/2.53, 10.86/6.379/2.53, 8.21/6.409/2.52</td>
</tr>
<tr>
<td></td>
<td>8.43/6.171/2.55, 11.58/6.098/2.51, 6.89/6.247/2.44</td>
</tr>
</tbody>
</table>
Fig. 4.1. Sample geometry and loading conditions.
Fig. 4.2. Schematic of the mesh configurations, (a) for the used CT samples, and (b) for an ideal CT sample without the mechanically loading slot.
Fig. 4.3. The values of $\sqrt{2\pi x} \sigma_{yy}(x, y = 0)$ and $\sqrt{2\pi x} E_x(x, y = 0)$ as a function of the distance from the crack tip.

Fig. 4.4(a). The critical load at fracture versus the notch length under purely mechanical loading.
Fig. 4.4(b). The mechanical fracture toughness versus the notch length.

Fig. 4.5. Typical optical microscopic picture of the fractured CT sample under purely mechanical or purely electrical loading, showing the two-dimensional failure behavior.
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Fig. 4.6(a). The critical voltage at fracture versus the notch length under purely negative electrical loading.

Fig. 4.6(b). The electric fracture toughness versus the notch length under purely negative electrical loading.
Fig. 4.7(a). The critical voltage at fracture versus the notch length under purely positive electrical loading.

Fig. 4.7(b). The electric fracture toughness versus the notch length under purely positively electrical loading.
Fig. 4.8(a). Experimental results for the failure of electrically conductive cracks in the poled PZT-8 ceramics under combined mechanical and negative electrical loading.
Fig. 4.8(b). Experimental results for the failure of electrically conductive cracks in the poled PZT-8 ceramics under combined mechanical and positive electrical loading.
Fig. 4.9. Fractographs of the poled PZT-8 ceramic samples under various combinations of electrical and mechanical loading, (a) under a purely mechanical load of $K_{a,C}^e = 1.25 \text{ MPa} \sqrt{\text{m}}$, (b) under combined mechanical and electrical loads of $K_{a,C}^e = 1.21 \text{ MPa} \sqrt{\text{m}}$ and $K_{e,C}^e = -92.8 \text{ kV} \sqrt{\text{m}}$, (c) under combined mechanical and electrical loads of $K_{a,C}^e = 0.43 \text{ MPa} \sqrt{\text{m}}$ and $K_{e,C}^e = -135.0 \text{ kV} \sqrt{\text{m}}$, and (d) under a purely electrical load of $K_{e,C}^e = -170.8 \text{ kV} \sqrt{\text{m}}$, where the arrow indicates the crack propagation direction and the arrow tip is located at the notch front.
Fig. 4.10 Schematics of experiment setup
Fig. 4.11 Dielectric breakdown image
Chapter Five

Application of the Concepts of Fracture Mechanics to the Failure of Conductive Cracks in Piezoelectric Ceramics

5.1. Introduction:

Piezoelectric ceramics have become preferred materials for a wide variety of electronic and mechatronic devices due to their pronounced dielectric, piezoelectric, and pyroelectric properties. Aging, fatigue and electrical and/or mechanical breakdown of the materials cause device failures, which are considered seriously in the design of piezoelectric devices. Piezoelectric ceramics are brittle and susceptible to cracking at all scales from electric domains to electronic devices. Because of the importance of electrical and mechanical reliability of these devices, there has been tremendous interest in studying the fracture behavior of such materials [1-6]. Zhang et al. [7] provide an overview on fracture of piezoelectric ceramics with summarizing current knowledge of the fracture of piezoelectric ceramics. Their attention is confined to fracture mechanics studies, yet experimental results are also examined for comparison with theoretical predictions. Later, Zhang and Gao [8] reviewed new development in the research on fracture of piezoelectric ceramics and introduced two newly developed models, i.e., the dielectric breakdown model and the charge-free zone model.
Interval electrodes have widely been adopted in electronic and electromechanical devices made of piezoelectric ceramics. These embedded electrodes may naturally function as pre-conductive cracks or notches if the Young's modulus of the electrode is much smaller than the Young's modulus of the ceramics. In addition, dielectric breakdown and partial discharge may convert an originally electrically insulating crack to an electrically conductive crack. Figure 3.3 schematically shows the similarity between a conductive crack under electrical loading and a conventional crack under mechanical loading. To ensure that the electric field inside the conductive crack remains zero, electric charges in the conductive crack surfaces must rearrange themselves to produce an induced field that has the same magnitude as the applied one but with the opposite sign. As a result, the charges in the upper and lower crack surfaces near the crack tip have the same sign, as shown in Fig. 3.3. The charges with the same sign repel each other and then have a tendency to propagate the crack. Garboczi [9] studied the contour-independent J-integral, which is a fundamental concept in fracture mechanics, for conductive cracks in dielectric materials. The J-integral for a conductive crack in a dielectric material under purely electric loading is similar to the J-integral for a conventional crack under purely mechanical loading, which is shown in Fig. 3.3 as well. It is therefore of practical importance and academic significance to apply the concepts of fracture mechanics to the failure of conductive cracks in piezoelectric and dielectric ceramics.

In 2003, Zhang et al. [10] proposed the Charge-Free Zone (CFZ) model by analogy with the Dislocation-Free Zone (DFZ) in order to understand the failure behavior of conductive cracks in dielectric ceramics under combined electric and mechanical loading. The CFZ model treats dielectric ceramics as mechanically brittle and
electrically ductile. Charge emission and charge trapping consume more work and thus lead to the electric ductility. Recent experimental observations on dielectric ceramics like silicon dioxide and semi-conductor materials like gallium nitride based on the novel technique electrostatic force microscopy (EFM) reveal that charge carrier can be trapped at the grain boundaries, interfacial surfaces and dislocations [17-21].

In the CFZ model, the local intensity factor of electric displacement has a non-zero value and consequently there is a non-zero local electric energy release rate, which contributes to the driving force to propagate the conductive crack. The merit of the CFZ model, similar to the DFZ model, lies in the ability to directly apply the Griffith criterion to link the local energy release rate to the fracture toughness in a completely brittle manner. As a result, an explicit failure criterion is resulted from the CFZ model to predict the failure behavior of conductive cracks in dielectric ceramics under combined electrical and mechanical loading. The significance of the failure criterion is that it provides designers of electronic and electromechanical devices with the electrical fracture toughness and the mechanical fracture toughness. The advantage of applying the failure criterion lies in the ability to predict the critical electric field and the critical mechanical load at which a dielectric ceramic material containing a conductive crack or an internal electrode fails under the combined electrical and mechanical loading. The critical electric field and the critical mechanical load are functions of the crack dimension or the length of the electrode, while the electrical fracture toughness and the mechanical fracture toughness are both material properties. Thus, one can predict the critical electric field and the
critical mechanical load when information on the sample geometry and on the
electrical and mechanical fracture toughness is available.

The CFZ model has been extended to predict the failure behavior of conductive
creaks in piezoelectric ceramics under electrical and/or mechanical loading [11].
Again, piezoelectric ceramics are treated mechanically brittle and electrically ductile
in the CFZ model such that charge emission and charge trapping are assumed to
occur at the conductive crack tip. The trapped charges partially shield the crack tip
from applied electrical field and the local electric intensity factor has a non-zero
value. Consequently, a non-zero local electric energy release rate contributes to the
driving force to propagate the conductive crack. The CFZ model yields an explicit
failure criterion to predict the failure behavior of conductive cracks in piezoelectric
ceramics under electrical and/or mechanical loading. Mathematically, the failure
formula takes an elliptic shape in terms of the normalized electric intensity factor and
the normalized stress intensity factor. When the normalized stress intensity factor is
set as the horizontal axis, the major semi-axis of the elliptic shape is rotated
anticlockwise 45° if applied electric fields are parallel to the poling direction, while
the major semi-axis of the elliptic shape is rotated clockwise 45° if applied electric
fields are anti-parallel to the poling direction. For dielectric materials, the failure
formula is reduced to a quarter of unit circle [10]. Zhang et al. [12] conducted
fracture experiments on electrically conductive cracks (deep notches) in poled lead
zirconate titanate PZT-8 ceramics under mechanical and/or electrical loading. The
experimental results revealed that the failure behavior of the conductive cracks in the
ceramics was described by an elliptic function of the normalized electric intensity
factor versus the normalized stress intensity factor under combined mechanical and
electric loading, thereby verifying the theoretical predictions from the CFZ model. However, the experimentally determined coupling factor deviated from the theoretical predicted value. This quantitative inconsistency encourages us to re-examine the CFZ model. In addition to reanalyze the experimental data on poled lead zirconate titanate PZT-8 ceramics, we conducted more tests on poled lead zirconate titanate PZT-4 ceramics and reported the experimental results here.

5.2. The Charge-Free Zone Model:

We briefly introduce the CFZ model here for the further development. The CFZ model is two-dimensional, in which charges are treated as line charges per unit length. Considering the image force and the driving force acting a line charge emitted from a conductive crack tip, there must be a charge-free zone in front of the crack tip for line charges to be continuously emitted from the crack tip. When more and more charges are emitted from the crack tip, these charges will entrap in the region of $ba$, as shown in Figure 3.2, where $ob$ denotes the CFZ size. If we define $f(x')$ to be the line charge number distribution function, the charge number located at $x'$ in the interval $dx'$ is $f(x')dx'$. The equilibrium condition that the electrical field, $E_x$, equals the critical value, $E_c$, in the charge trap zone is described by

$$
\frac{K_e}{\sqrt{2\pi}} + Q \int_{b}^{a} \frac{f(x')\sqrt{x'}}{\sqrt{x(x-x')}} dx' = E_c, \quad b \leq x \leq a,
$$

(1)
where $K^o_e$ is the applied electric intensity factor, $E_c$ is a critical stress field, about which the charge lines are mobile and below which the charge lines cannot move [13], and

$$Q = \frac{q}{2\pi\kappa_e},$$

$q$ stands for a line charge per unit length, and $\kappa_e$ denotes an equivalent dielectric constant. The first term on the left side in Eq. (1) represents the applied electric field, while the second term on the left side stands for the electric field induced by the electric charges. Solving the integral equation and using the local energy release rate as the failure criterion [11], the CFZ model gives the following failure formula:

$$\left(\frac{K^o_{\sigma,C}}{K^o_{\sigma,C}}\right)^2 + \eta \left(\frac{K^o_{\sigma,C}}{K^o_{\sigma,C}}\right) \left(\frac{K^o_{E,C}}{K^o_{E,C}}\right) + \left(\frac{K^o_{E,C}}{K^o_{E,C}}\right)^2 = 1,$$

where the subscript "C" denotes fracture or failure, $K^o_{\sigma,C}$ is the fracture toughness in terms of the critical stress intensity factor under purely mechanical loading, $K^o_{E,C}$ is the electric fracture toughness in terms of the electric intensity under purely electrical loading, and $\eta$ is called the coupling factor mentioned above. Mathematically, Eq. (3) has the form of $x^2 + \eta xy + y^2 = 1$ with $x = K^o_{\sigma,C} / K^o_{\sigma,C}$ and $y = K^o_{E,C} / K^o_{E,C}$. The mathematic equation can be expressed in the standard form of an ellipse,
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\[ \hat{x}^2 / [2/(2 + \eta)] + \hat{y}^2 / [2/(2 - \eta)] = 1 \] , where the \((\hat{x}, \hat{y})\) coordinate system is established by rotating 45° from the horizontal axis of the \((x, y)\) coordinate system.

The absolute value of \(\eta\) is less than two and thus Eq. (3) indeed describes an ellipse in terms of the normalized applied intensity factors. For dielectric materials, the piezoelectric constants are zero and thus the coupling factor is zero. Therefore, the interaction term, i.e., the second term on the left hand-side of Eq. (3) disappears, thereby reducing Eq. (3) to the failure criterion for conductive cracks in dielectric materials [10].

If the conductive crack is parallel to the poling direction, the coupling factor is given by

\[
\eta = \sqrt{\frac{2e}{\left(\varepsilon^2 + \kappa M d^2\right)^{1/2}}},
\]

where \(\varepsilon = \Lambda_2(1 - \Omega) - \Omega \hat{H}_{24} / \hat{H}_{22}, \quad M = 2 \hat{H}_{22}, \quad \text{and} \quad \kappa = \left[ \hat{H}_{44} + \left( \hat{H}_{24}^2 / \hat{H}_{22} \right) \right] / 2 \)

are called the effective piezoelectric constant, the effective elastic modulus, and the effective dielectric constant, respectively, the parameter \(\Omega\) is defined as the ratio of the local electric intensity factor to the applied, i.e., \(\kappa^i = \Omega K^i\), which represents the shielding level of the trapped charges to the conductive crack tip, the constants, \(\Lambda_2\), \(\hat{H}_{22}, \hat{H}_{24}\) and \(\hat{H}_{44}\) are material constants related to the orientation of the tested sample [11, 12]. The sign of \(\eta\) is determined by the polarity direction of an applied electric field. An applied electric field is defined to be positive if the applied field is
parallel to the crack propagation direction, whereas an applied electric field is
defined to be negative if the applied field is anti-parallel to the crack propagation
direction. For the poling direction being parallel to the crack propagation direction,
the $\eta$ parameter take the positive sign under negative electric loading, while the $\eta$
parameter takes the negative sign under positive electric loading. For the poling
direction being anti-parallel to the crack propagation direction, however, the $\eta$
parameter take the negative sign under negative electric loading, while the $\eta$
parameter takes the positive sign under positive electric loading [11, 12]. Under
purely mechanical mode I loading, the critical energy release rate is determined from

$$
\Gamma = \frac{\left( K_{\sigma,c}^\sigma \right)^2}{2M}.
$$

(5)

In the CFZ model, the critical value of the local energy release rate is taken as the
failure criterion, which gives the relationship between the fracture toughness under
purely mechanical loading and the electric fracture toughness under purely
mechanical loading as [11, 12]

$$
\mathcal{E} = \frac{\left( K_{\sigma,c}^\sigma \right)^2}{\left( K_{\kappa,c}^\kappa \right)^2} \text{ with } \mathcal{E} = e^2 + \kappa M \Omega^2.
$$

(6)

Eq. (6) will be used also in the analysis of the experimental data.

5.3. Piezoelectric constants or moduli:
Piezoelectric constants, $e$, are a three rank tensor, which is related to the piezoelectric modulus tensor, $d$, by $d = es$, where $s$ (italic) denotes the elastic compliance tensor. It is convenient to experimentally measure piezoelectric moduli. For example, Fig. 5.1 illustrates the so-called butterfly curve, where mechanical strain is measured under an applied electric field. The slope of strain versus electric field is defined as piezoelectric modulus and usually the slope at zero electric field is regarded as the piezoelectric modulus, which is treated as a constant under low electric fields such that mechanical strain is linearly proportional to electric field. As the butterfly curve indicates that when electric field strength is large, the strain, $\varepsilon$, varies nonlinearly with the electric field strength, $E$. In the vicinity of a conductive crack tip, the electric field has a $\sqrt{r}$ singularity, where $r$ denotes the distance from the crack tip, based on the analysis of linear electroelastic fracture mechanics. If we use the conventional value of piezoelectric modulus or piezoelectric constant in the analysis, we shall obviously overestimate the piezoelectric effect. Frankly speaking, nonlinear approach is needed to understand the nonlinear failure behavior of piezoelectric ceramics under combined mechanical and electric loading. However, nonlinear analytic approach is extremely challenging. That is why the CFZ model is proposed to solve the nonlinear problem in a feasible manner, which converts a nonlinear approach to be, in some extent, linear. In such a sense, we would like to introduce a secant piezoelectric modulus or constant, which is defined as the slope of a secant drawn from the origin to a given point of the $s$ versus $E$ curve. Schematic illustration of secant piezoelectric modulus is given in Fig. 5.2. The great advantage of employing secant piezoelectric modulus lies in that the linear constitutive equation can be still used in the analysis, while the overestimation of the piezoelectric effect can be reduced, because the strain induced by final maximum value of an electric
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field is exactly calculated by the linear constitutive equation. Fig. 5.2 shows three secant piezoelectric moduli, which are represented by three tangential functions of angles $\theta_1$, $\theta_2$ and $\theta_3$. Under low electric field, the strain is linearly proportional to the electric field and hence the secant piezoelectric modulus is reduced to the nominal piezoelectric modulus, as shown in Fig. 5.2. As the maximum value of an applied electric field increases, the secant piezoelectric modulus is greatly decreased. Note that under an extremely high electric field, all electric domains will align themselves along the electric field direction and polarization and strain will reach their saturated values, respectively. After that, further increasing electric field will increase strain slightly, thereby indicating the secant piezoelectric modulus decreases further and could theoretically approach zero.

5.4. Numerical calculations:

Material properties of PZT-4 and PZT-8 piezoelectric ceramics are given in Tables 4.1 and 4.2, respectively.

Table 1 Material properties of poled PZT 4

<table>
<thead>
<tr>
<th>Elastic Constants $c$ ($10^{10}$ N/m²)</th>
<th>Piezoelectric Constants $e$ (C/m²)</th>
<th>Dielectric Constants $\kappa$ (10^{-9} F/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c_{11} = 13.9$,</td>
<td>$e_{31} = -6.89$</td>
<td>$\kappa_{11} = 6.0$</td>
</tr>
<tr>
<td>$c_{13} = c_{12} = 7.43$, $c_{33} = 11.3$</td>
<td>$e_{33} = 13.84$</td>
<td>$\kappa_{33} = 5.47$</td>
</tr>
<tr>
<td>$c_{44} = 2.56$</td>
<td>$e_{35} = 13.44$</td>
<td>$\kappa_{22} = 6.0$</td>
</tr>
</tbody>
</table>
Table 4.2 Material properties of poled PZT 8

<table>
<thead>
<tr>
<th>Elastic Constants $c$ ($10^{10} N/m^2$)</th>
<th>Piezoelectric Constants $e$ ($C/m^2$)</th>
<th>Dielectric Constants $\kappa / \kappa_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c_{11} = 14.9, c_{33} = 13.2$</td>
<td>$e_{31} = -4.1$</td>
<td>$\kappa_{11} = \kappa_{22} = 1290$</td>
</tr>
<tr>
<td>$c_{44} = 3.13$</td>
<td>$e_{33} = 13.2$</td>
<td>$\kappa_{33} = 1000$</td>
</tr>
<tr>
<td>$c_{12} = c_{13} = 8.11$</td>
<td>$e_{15} = 10.3$</td>
<td>$\kappa_0 = 8.855 \times 10^{-12} \text{Cm}^2/\text{V}$</td>
</tr>
</tbody>
</table>

The secant piezoelectric constants are approximately defined as $\alpha e_{ij}$ with $\alpha$ changing from zero to one. To have a smooth mathematical calculations, we took new units or scales marked with (*) by letting $c_{ijkl}^* = 10^{-9} c_{ijkl}$ and $\kappa_{ij}^* = 10^9 \kappa_{ij}$ and other properties and constants having the same units or scales as the original ones [12]. Following the matrix operation given in Reference [11, 12], we calculate values of the constants, $A_{ij}$, $\tilde{H}_{22}$, $\tilde{H}_{24}$ and $\tilde{H}_{44}$, and then the effective dielectric constant and the effective elastic modulus. Figure 5.3 shows the effective dielectric constant, $\kappa$, and the effective elastic modulus, $M$, as functions of $\alpha$. As expected, both the effective dielectric constant and the effective elastic modulus are not sensitive to the $\alpha$ value. We may take the results for PZT-8 ceramics as an example. The values of $M$ and $\kappa$ decrease from 50.73 to 49.19 ($\times 10^{10} \text{ N/m}^2$) and from 12.17 to 10.04 ($\times 10^{-9} \text{ F/m}$), respectively, as the $\alpha$ value decreases from one to zero.

As described above, the effective piezoelectric constant, $e$, depends on both $\alpha$ and $\Omega$. Figure 5.4 illustrates the effective piezoelectric constant as a function of $\alpha$ for $\Omega = 0.1, 0.2, 0.3, 0.4$ and $0.45$. As expected again, Fig. 5.4 shows that the effective piezoelectric constant is zero if $\alpha = 0$, no matter how large is the value of $\Omega$. If the
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\(\alpha\) parameter has a nonzero value, the effective piezoelectric constant is almost linearly proportional to the value of \(\alpha\). For a given value of \(\alpha\), the effective piezoelectric constant increases as the \(\Omega\) parameter decreases.

Figure 5.5 shows the \(\eta\) parameter as a function of \(\alpha\) for \(\Omega=0.1\), 0.2, 0.3, 0.4 and 0.45. As described above the failure formula will be reduced to a quarter of circle if the \(\eta\) parameter is zero. When \(\alpha=0\), the \(\eta\) parameter is zero, no matter how large is the value of \(\Omega\), as shown in Figure 5.5. When the \(\alpha\) parameter has a nonzero value, the \(\eta\) parameter is larger if the value of \(\Omega\) is smaller. For \(\Omega=0.45\), the \(\eta\) parameter approximately increases linearly with the increase of \(\alpha\). For a small value of \(\Omega\), the \(\eta\) parameter increases nonlinearly with the increase of \(\alpha\). For example, if \(\Omega=0.1\), the \(\eta\) parameter increases very quickly as \(\alpha\) increases from zero to about 0.25. Then, the \(\eta\) parameter increases much slowly as \(\alpha\) increases from 0.25 to unity. Of course, the behavior of the \(\eta\) parameter depends on the material properties. That is why the curves for PZT-4 ceramics are different from the curves for PZT-8 ceramics.

5.5. Experimental procedure:

Pre-notched Compact Tension (CT) samples, as shown in Fig. 5.6, were made from commercial available piezoelectric ceramics PZT-4 and PZT-8 (Morgan ElectroCeramics). The sample preparation for PZT-4 ceramics is the same as that for PZT-8 ceramics [12]. It should be pointed out that the purchased PZT-4 and PZT-8 ceramics are poled by the manufacturer and all the material data are provided by the manufacturer. After preparing the fracture samples, we did not re-poling the samples.
and we did not measure the material constants of the pre-notched CT samples either, although the sample preparation might cause changes in the material constants.

The experimental procedure was described in the previous publication [10, 12]. The tests on the deeply notched conductive samples were conducted under purely mechanical loading, purely electric loading, and combined mechanical and electric loading. The critical voltage and critical mechanical load at failure were recorded and used to calculate the electric intensity factor and the stress intensity factor.

Finite Element Analysis (FEA) was also carried out to calculate stress intensity factor and electric intensity factor of the used samples. In the finite element analysis, the material constants listed in Tables 1 and 2 were used. The FEA details were described in the previous publications [10, 12]. The FEA results for the PZT-8 ceramics are reported in Reference [12]. In the following, we report the FEA results for the PZT-4 ceramics.

For purely elastic bodies, it is well known that the value of stress intensity factor is determined only by the load level and the sample geometry and independent of elastic constants. For piezoelectric ceramics, the FEA results indicate that stress intensity factor, $K_\sigma$, and electric intensity factor, $K_e$, are slightly related to the material constants. Furthermore, a mechanical load may induce an electric intensity factor and an electric voltage may produce a stress intensity factor due to the coupling behavior of piezoelectric ceramics [12, 13]. Under the testing conditions, however, the coupling-induced electric intensity factor and the coupling-induced stress intensity factor are correspondingly much smaller than the electric intensity.
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factor directly produced by a voltage and the stress intensity factor directly produced by a mechanical load. Therefore, the coupling-induced electric intensity factor and the coupling-induced stress intensity factor are ignored in the present study. With this approximation, the AEF yields the following equations:

\[ K_\sigma = \frac{P}{B\sqrt{w}} f_1(t), \quad (7a) \]

\[ K_E = \frac{V}{\sqrt{w}} f_2(t), \quad (7b) \]

where \( P \) and \( V \) denote a mechanical load and an electric voltage, respectively, \( B \) is the sample thickness, \( w \) is the sample length along the notch (or crack) direction, \( t = (w - l)/w \) is the normalized notch length, as shown in Fig. 5.6, and \( f_1(t) \) and \( f_2(t) \) are given by,

\[ f_1(t) = -5.60794 + 66.515 t - 43.3734 t^2 - 139.197 t^3 + 226.173 t^4, \quad (7c) \]

\[ f_2(t) = 10.5001 - 81.4567 t + 254.772 t^2 - 338.301 t^3 + 161.791 t^4, \quad (7d) \]

respectively. Here, \( w = 9 \) mm was used in the AFE and the normalized notch length, \( t \), is within a range from 0.33 to 0.72.

5.6. Results and discussion:
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For the PZT-8 ceramics, the experimental results were published in Reference [12], which will not be repeated here. In the following, we report the experimental results for the PZT-4 ceramics.

Using Eq. (7) and the critical loads at fracture, we calculate the critical stress intensity factors under purely mechanical loading. The mean and standard deviation of the critical stress intensity factor is \( K_{\sigma,c}^o = 0.934 \pm 0.063 \text{ MPa}\sqrt{m} \) under purely mechanical loading. The relative error in the experimental data is about \( \pm 6.6\% \). The value of \( K_{\sigma,c}^o = 0.934 \text{ MPa}\sqrt{m} \) is almost the same as the value determined by Fu et al. [14] and is smaller than the fracture toughness, \( K_{\sigma,c}^o = 1.26 \text{ MPa}\sqrt{m} \), of the poled PZT-8 ceramics [12], which might be attributed to the different \( w \) value used in the FEA, which is uncertain by about 1 mm due to the mechanical loading slot. For the PZT-4 ceramics, \( w = 9 \) mm was used, while \( w = 10 \) mm was used for the PZT-8 ceramics. If the size of CT samples is big, this uncertain will be reduced greatly.

An applied electric field could fracture the poled ceramic samples with conductive deep and sharp notches [14, 15]. The fracture was accompanied with dielectric discharging. As described above, the charges in the upper and lower surfaces of a conductive crack have the same sign. The charges with the same sign repel each other and generate a Coulombic force that opens and propagates the conductive crack. For the CT samples of the brittle ceramics, the fracture process is unstable because the stress and electric intensity factors increase both monotonically with the crack length, which means that failure will occur once the crack propagation is triggered mechanically and/or electrically. The fracture process and the dielectric breakdown
may occur simultaneously in the CT samples, even so charge emission from the conductive notch (crack) tip may happen prior to the process of fracture and dielectric breakdown. It is the sample geometry and the loading condition that makes the failure become two-dimensional. The experimental results all reveal the two-dimensional failure behavior, as shown in Fig. 5.7. In the present work, we study only the onset of the failure process and the critical values of the voltage and the mechanical load at the onset are used to calculate the electrical fracture toughness and the mechanical fracture toughness.

Using Eq. (7) and the critical voltages, we calculate the critical electric intensity factors under purely electric loading. The mean and standard deviation of electric fracture toughness are $K_{\varepsilon,C}^e = 280.5 \pm 13.5 \, kV / \sqrt{m}$ and $K_{\varepsilon,C}^c = -342.5 \pm 15.5 \, kV / \sqrt{m}$ for the poled PZT-4 ceramics under purely positive and negative electrical loadings, respectively. Like the mechanical fracture toughness, the electric fracture toughness is a material constant independent of the crack length and thus can serve as a failure criterion for conductive cracks (or deep notches) in piezoelectric ceramics under purely electrical loading. The relative errors in the experimental data of the electric fracture toughness are about $\pm 4.8\%$ under purely positive electrical loading and about $\pm 4.5\%$ under purely negative electrical loading, which are comparable to that of the mechanical fracture toughness of about $\pm 6.6\%$. The experimental errors may be attributed to the following reasons: 1) the notch is not sharp enough, and 2) there are many defects such as grain boundaries and pores in the PZT ceramics fabricated by sintering PZT powders, etc.
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To compare the electrical fracture toughness with the mechanical fracture toughness, we convert them correspondingly to the critical values of the energy release rates. The mean value of the critical energy release rate is $G_{e,C} = 452 \pm 42 \, N/m$ under purely positive electric loading and $G_{e,C} = 673 \pm 61 \, N/m$ under purely negative electric loading, whereas the mean value of the critical energy release rate is $G_{\sigma,C} = 8.7 \pm 0.4 \, N/m$ under purely mechanical loading. The electrical toughness under either purely positive electric loading or purely negative electric loading is over 50 times higher in magnitude than the mechanical toughness. The high electrical toughness is attributed to electrical plastic deformation, such as electrical discharge, etc., that occurs at the tip of the conductive notch, forming an electrical plastic zone. Actually, dielectric breakdown accompanies the fracture under purely electrical loading. The fracture surfaces are flat for samples fractured under purely mechanical loading, while purely electrical loading yields rough fracture surfaces, as shown in Fig. 5.7. In the electrically fractured samples, the discharge may locally burn the sample, as exhibited by the dark area in the fractograph in Fig. 5.7(b). It is the electrical plastic deformation that consumes more energy and thus leads to the high electrical fracture toughness. Furthermore, the mechanical toughness may be regarded as the critical value of the local energy release rate, $\Gamma$, as indicated by Eq. (5). If the local energy release rate is taken as a failure criterion, the experimental result that the electric toughness is much higher than the mechanical toughness gives a hint that a shielding mechanism must take place under electrical loading.

From the experimental results under purely electrical or mechanical loading, we estimate the value of the parameter, $\Xi = \varepsilon^* + \kappa M \Omega^2$, as indicated in Eq. (6), which
are $11.09 \ [N/(mV)]^2$ under purely positive electrical loading and $7.44 \ [N/(mV)]^2$ under purely negative electrical loading. As described above, the $\Xi$ parameter is a function of the $\alpha$ and $\Omega$ parameters. Therefore, for a given value of $\Xi$, the $\Omega$ parameter is a function of the $\alpha$ parameter.

The experimental data of critical loads and the critical voltages at fracture under combined mechanical and electrical loading are more scattering than those under purely mechanical or electrical loading. For instance, for 10 PZT-4 samples with the almost same notch length of 6.40 mm, the value of the mechanical load at fracture varies from 8.75 N to 18.1 N under a constant voltage of 7.0 kV. For another 13 PZT-4 samples with the almost the same notch length of $6.63 \pm 0.15$ mm, the voltage at fracture varies from 6700 to 13100 V under a constant force of 9.0 N. The phenomenon that combined mechanical and electrical loading leads to an increase in the experimental error has been observed before in the bending tests on PZT-841 ceramics [7, 16] and in the fracture tests on depoled PZT-4 ceramics [10] and poled PZT-8 ceramics [12]. In the bending tests on the PZT-841 ceramics, the mechanical bending strength at an applied electric field of 10 kV/cm varies from about 30 MPa to about 120 MPa with a relative error of about 33% [7, 16]. In the fracture tests on the depoled PZT-4 ceramics under a constant voltage of 3.0 kV [10], the value of the mechanical load varies from 18.4 N to 29.0 N for the 17 samples with the almost same notch length of $5.04 \pm 0.19$ mm. For the poled PZT-8 ceramics [12], the value of the mechanical load varies from 21.8 N to 29.3 N under a constant voltage of 2.0 kV for the 9 samples with the almost same notch length of $4.64 \pm 0.05$ mm. Due to the large scattering in the experimental data, we tested sufficiently large number of samples to ensure statistical assessment of the failure behavior. From the
experimental data and the crack length, we calculate the critical stress intensity factor, $K^{\sigma}_{s,C}$, and the critical electric intensity factor, $K^{\sigma}_{E,C}$, for each sample. Then, we normalize the critical stress intensity factor by the critical stress intensity factor under purely mechanical loading, and normalize the critical electric intensity factor by the critical electric intensity factor under purely electric loading. Figures 5.8 and 4.9(b) show the relationships of the normalized electric intensity factor versus the normalized stress intensity factor under combined mechanical and electrical loading for positive and negative electric fields, respectively. As described above, the CüZ model predicts that the failure behavior should have a shape of an ellipse. We adopt the orthogonal distance fitting method to fit the experimental data with the mathematic equation, $x^2 + \eta xy + y^2 = 1$. The fitting results are illustrated by the dashed curves in Fig. 5.8 and Fig 5.9, where a quarter of unit circle is plotted by solid curves for comparison. As described above and in Reference [12], when the poling direction is anti-parallel to the crack propagation direction, the $\eta$ parameter take the positive (or negative) sign under positive (or negative) electric loading. The fitting results indicate that $\eta=0.03$ and the minor semi-axis of the ellipse is on the line of $K^{\sigma}_{s,C}/K^{\sigma}_{E,C} = K^{\sigma}_{s,C}/K^{\sigma}_{E,C}$ under positive electrical loading, whereas $\eta=0.18$ and the major semi-axis of the ellipse is on the line of $K^{\sigma}_{s,C}/K^{\sigma}_{E,C} = K^{\sigma}_{s,C}/K^{\sigma}_{E,C}$ under negative electrical loading. As plotted in Fig. 5.5 and Eq. (4), the $\eta$ parameter is a function of the $\alpha$ and $\Omega$ parameters. For a given $\eta$ parameter, the $\Omega$ parameter can be expressed in terms of the $\alpha$ parameter. Since both $\eta$ and $\Omega$ parameters can be determined experimentally, simultaneously solving Eqs. (4) and (6) gives the values of $\alpha$ and $\Omega$. Fig. 5.10 shows the $\Omega$ parameter as a function of $\alpha$ parameter for a given value of $\eta=0.03$ or $\eta=0.18$ and for a given value of $\Xi=11.09$.
\[(N/(mV))^2 \text{ or } \xi = 7.44 \times (N/(mV))^2 \text{ for the PZT-4 ceramics, in which the cross point yields the values of } \alpha = 0.05 \text{ and } \omega = 0.17 \text{ under combined mechanical and positive electrical loading and } \alpha = 0.01 \text{ and } \omega = 0.21 \text{ under combined mechanical and negative electrical loading.}

Similarly, we re-analyze the failure data of conductive cracks in PZT-8 ceramics under combined mechanical and electric loading. Figures 4.8 (b) and 4.8(a) illustrate the normalized failure data under combined loading with positive and negative electric fields, respectively. The experimental results for the PZT-8 ceramics give that \(\xi = 58.95 \times (N/(mV))^2 \text{ and } \eta = 0.52 \text{ for positive electrical loading and } \xi = 58.24 \times (N/(mV))^2 \text{ and } \eta = -0.16 \text{ for negative electrical loading. The values of } \alpha \text{ and } \omega \text{ are also determined with plots to be } \alpha = 0.76 \text{ and } \omega = 0.31 \text{ under combined mechanical and positive electrical loading and } \alpha = 0.25 \text{ and } \omega = 0.34 \text{ under combined mechanical and negative electrical loading, as shown in Fig. 5.11. Comparing the results for the PZT-8 ceramics with those results for PZT-4 ceramics indicates that both } \xi \text{ and } \omega \text{ parameters for the PZT-4 ceramics are correspondingly smaller than these for the PZT-8 ceramics. One reason for this difference is that the electric fracture toughness for the PZT-4 ceramics is much higher than that for the PZT-8 under purely positive or negative electric loading. The electric fracture toughnesses for the PZT-4 ceramics under purely positive and negative electric loading are } K_{e,c}^o = 280.5 \pm 13.5 \text{ kV}/\sqrt{m} \text{ and } K_{e,c}^o = -342.5 \pm 15.5 \text{ kV}/\sqrt{m} \text{, respectively, while for the PZT-8 ceramics, they are } K_{c,c}^o = 157.1 \pm 14.1 \text{ kV}/\sqrt{m} \text{ and }
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\[ K^0_{e,c} = -165.2 \pm 12.7 \, kV / \sqrt{m} \] [12]. At the moment, the reason for the big difference in the electric fracture toughness of the PZT-4 and PZT-8 ceramics is still unclear.

5.7 Concluding remarks:

The concept of secant piezoelectric constant is introduced in this work to refine the CFZ model. With the use of secant piezoelectric constant, the experimental results can be well explained by the CFZ model. The theoretical modification and the reanalysis of the experimental data indicate that the role of the piezoelectric effect in the failure of conductive cracks in piezoelectric ceramics could be reduced due to the high electric field at the crack tip. The empirical fitting of the experimental results for the PZT-4 and PZT-8 ceramics under positive and negative electric loading yields the \( \eta \) parameters smaller than the corresponding values from the prediction of the CFZ model with the nominal piezoelectric constants, thereby strongly supporting the rationale of using the secant piezoelectric constant. Although the absolute values of the experimental data exhibit great scattering and big difference between the PZT-4 and PZT-8 ceramics, the normalized experimental results can be predicted by the modified CFZ model. As described in the previous publications [10, 11], the CFZ model is two-dimensional, in which the charges are treated as line charges per unit length and all charges are allocated along a strip. The present work indicates that the two-dimensional CFZ model is able to grab the physical nature of the three-dimensional phenomena of electric charges emitted from a conductive crack tip and trapped in front of the tip. The distinctive advantage of the two-dimensional CFZ
model lies in that it is sample and the failure formula derived is easy to apply to engineering practice.
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References:


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Fig. 5.1 Schematic plot of a butterfly loop of mechanical strain and electric field strength.
Fig. 5.2 Schematic plot of secant piezoelectric modulus
Fig. 5.3 The effective dielectric constant, $\kappa$, and the effective elastic modulus, $M$, as functions of the $\alpha$ parameter.
Fig. 5.4 The effective piezoelectric constant, $e$, as a function of the $\alpha$ parameter.
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Fig. 5.5 The $\eta$ parameter as a function of the $\alpha$ parameter.
Fig. 5.6 Sample geometry and loading conditions for the PZT-4 ceramics.
Fig. 5.7 Optical pictures of the fractured samples, (a) front view and (b) fractograph, where mechanical and electrical mean purely mechanical and electrical loading, respectively. The dark area in the fractograph under purely electrical loading indicates that a burned area caused by dielectric discharging.
Fig. 5.8 Experimental results for the failure of electrically conductive cracks in the poled PZT4 ceramics under combined mechanical and positive electrical loading.
Fig. 5.9 Experimental results for the failure of electrically conductive cracks in the poled PZT4 ceramics under combined mechanical and negative electrical loading.
Fig. 5.10 The $\Omega$ parameter varies with the $\alpha$ parameter for a given $\eta$ parameter or a given $\Sigma$ parameter for the PZT-4 ceramics.
Fig. 5.11 The $\Omega$ parameter varies with the $\alpha$ parameter for a given $\eta$ parameter or a given $\Xi$ parameter for the PZT-8 ceramics.
Chapter Six

Electric breakdown in polymeric vinyl chloride (PVC)

6.1 Physics background about charge injection and charge trapping

Polymers are widely used as preferable insulating materials due to their extremely poor electric conductivity. Compared with good conductors, which are usually metals like silver, gold and copper, the conductivity of polymers can be over 20 orders in magnitude lower. Unlike most metallic materials, which are crystalline, polymeric materials can exist as amorphous materials, as crystalline materials, or as mixers of crystalline and amorphous materials. Polymers even with high crystallinity contain considerable amorphous parts. Polymers are relatively disordered materials when they are compared with metallic materials or semi-conductor materials. Polymers are molecular materials. In a molecular chain, the chemical bonds are covalent and the interactions are strong. But the intermolecular interactions are weak van der Waals forces.

For perfect crystalline materials, the energy states are periodical. Amorphous regions in a polymer change energy states of energy bands and create localized energy states. Typically they are: surface states induced by strain or chemical reaction, surface dipole states, bulk dipole states, bulk molecular ion states, impurities related to different chemical groups, polar groups and ionic groups, chain ends or dangling bonds, chain branches, chain folds, changes in tacticity or stereochemistry,
crystalline-amorphous boundaries, broken bonds, polaron states related to trapped charge and the region of surrounding polarized dielectric, and local density fluctuation [1]. In crystalline semiconductor materials or ceramics, defects like dislocation can also act as localized states [4]. In short, any local distortion in the chemical and/or electrical distribution may cause the localized states.

6.1.1. Charge injection

The Fermi energies of a metal electrode and a ceramic dielectric material are different. After an electrode contacts with a dielectric material, the electrons flow from the material with a higher Fermi energy to the material with lower Fermi energy until equilibrium is achieved. The energy band change is schematically shown in Fig.6.1. Electrons flow from the semiconductor to the metallic electrode and hence form an electron depletion area in the semiconductor near the contact surface. Fig.6.1 shows electron blocking contact. For most cases the contact is not ohmic contact. For ohmic contact, the relationship between current and applied voltage follows Ohm’s Law.

A biased voltage can reduce the potential barrier height as well as its width. Therefore, charge carriers can be more easily injected into the semiconductor or insulator as schematically shown in Fig.6.2. There are three ways in which energy required for charge carriers to escape from the metal may be supplied: (1) by thermal methods in a process known as thermionic emission, (2) by the application of high electric fields in field emission, and (3) by photon absorption at sufficiently short wavelengths in the photoemission process. Mechanisms (1) and (3) may be modified
by the Schottky effect which arises from the field-dependent lowering of the potential barrier to injection. As the biased potential increases, the width and height of the potential barrier both decrease. More extremely, if the Fermi energy difference is large and the applied field is sufficiently high, the width of the potential barrier can become so thin that quantum-mechanical tunneling occurs [3]. For such kind of cases, tunneling mechanism may become the dominant mechanism.

6.1.2 Charge trap

Disordered regions in dielectrics change the energy states and introduce some energy states as shown in Fig.6.4. Trap models are often interpreted in terms of a modified energy-band model. The model indicates that traps can be treated as the localized states belonging to certain molecules or molecular groups (see Fig.6.3), which are affected by their environment, are different in different regions of the material. The modified energy-band model is typically suitable for polymers since most polymers are amorphous or semi-crystalline. Even for polymers with a high degree of crystallinity, there are still considerable amorphous regions between the crystalline areas as schematically shown in Fig 6.7. The whole energy states for the polymers can be schematically described as Fig.6.5. The regular and periodical energy states, which can be well defined by energy band theory, are sandwiched (surrounded) by the randomly distributed energy states in the amorphous regions. Traps can be classified as deep or shallow types based on their energy states as shown in Fig.6.6. Trap energy states near the valence band for holes or near the conduction band for electrons are shallow traps. Trap energy states near the Fermi level are deep traps. Their distribution is described by the density of states as shown in Fig.6.3.
6.2. Introduction

In short, charge can be injected into the bulk and trapped at trapping sites, and then transfer from one site to another. Observations of charge carriers trapped at the trapping sites such as grain boundaries, dislocations and interfaces in ceramics or semi-conductor structures have been conducted using the electrostatic force microscopy technique (EFM) [4]. More experiments on the charge trapping phenomena have been done on polymeric materials. Trapped charges, also called space charges, can be transferred from one site to another under the external influences such as an electric field. The process corresponds to the local oxidation or local reduction. Initially Faraday pail method was used to measure the trapped charge type and distribution. When the charged object is put into a metallic Farady pail, based on the electrostatic induction, the charges in the object would induce same amount charges with different charge types on the inner and outer surfaces. This method needs shaving the electrodes away at low temperature to avoid any loss of the stored charge. Recently, a few techniques have been developed to measure nondestructively the space charge distribution in dielectrics, which include the thermal pulse, laser-intensity modulation, laser induced pressure pulse, thermoelectrically-generated laser-induced pressure pulse, pressure wave propagation, non-structured acoustic pulse, laser-generated acoustic pulse, acoustic probe, piezoelectrically generated-pressure step, electro-acoustic stress pulse, electroptic and spectroscopic method, pulsed electroacoustic method (PEA) and novel EFM technique. Each method has its own limit. A comprehensive review on the measurement methods was given in ref [5].
The study of charge carriers' behavior in polymers and their affects on failures of polymers has been the focus since their wide application in the high power industries. The study of charge carrier injection concerning polymeric materials attracts great attention now because the performance of the thin-film field-effect transistors based on organic materials as the active semi-conducting components has experienced impressive improvement in recent years [12, 13]. Previous experimental observations on polymeric materials like polyethylene [10, 14-15], polyester, Teflon [11], and polyvinylchloride (PVC) [16] were made in terms of emission processes. Recently, the PEA method becomes popular. With the PEA method, space charge distributions, space charge type [6-9], the current density for positive and negative polarity conditions, as well as the profile change of charge density distribution at different temperatures [9] were measured for various polyethylene (PE) [7] thin films under different electrical stresses. All the results of the studies on PE sandwiched with different electrodes may be summarized as this: positive charge carriers injected from anode and concentrated near the electrode had much higher mobility than the negative charge carriers injected from cathode [6-9].

A routine and widely accepted method to study the dielectric breakdown behavior of polymers was the so-called pin-plate method. A metallic rod with a sharp tip was inserted into a bulk polymeric material in the direction perpendicular to the opposite surface. The metallic rod and the opposite surface acted as electrodes. Experimental researches on PE showed that polarity not only had great influence on the tree initiation voltage and the breakdown voltage [17], but also had great influence on the
trees' shape near the tip [18]. For negative polarity, the tree initiation and breakdown voltages were much lower and the tree was sparse short bush like; for positive polarity case, long single path trace initiated from the tip [18].

The advantage of this measurement method was its simplicity. However, the key measurement parameter, the breakdown (DB) voltage, had only a vague physics meaning. In addition, the measurement results were sensitive to the tip shape and the interface conditions. In the present study, the compact tensile (CT) test method, which had been well applied in the study of the fracture behavior of poled ceramics PZT 4 and PZT 8, and the depoled PZT 4 [19-22], was adopted to investigate the electrical fracture behavior of PVC. The concept of electrical fracture toughness, which was more physically meaningful, was used to characterize the electrical properties of polymer.

In this work, CT samples and double notched samples of PVC were prepared to carry out the electrical failure study as well as the possible influence of polarity on the electrical fracture behavior of PVC.

6.3 Experiment

PVC used here was semi-crystalline. All samples had the same thickness of 3 mm. The dimensions of CT samples and double notched CT samples were, respectively, 20 mm (Length) X 20 mm (Width) and 22 mm (Length) X 20 mm (Width), as shown in Fig.6.8. Notches were cut with diamond wheel and then sharpened with a thin blade to make the notch tips sharp. Then samples were ultrasonically cleaned in de-
ionized (DI) water. Silver paint was filled into the notch repeatedly until notches were fulfilled. The fulfillment quality of the samples was later checked with an optical microscope in case tiny air gap might exist near the notch tip area. Silver paint was also spread along two surfaces to function as electrodes. Finally, copper wire, used to connect with the anode and cathode of power supply, was soldered by a mixer of silver power and epoxy. Electrodes were then solidified at 40°C for a few hours.

Experiment setup was schematically shown in Fig.4.10. The power supply used in this work provided with a negative high voltage and its anode was grounded for safety. To avoid any potential damage to the digital oscilloscope by the high voltage, two attenuators with a fixed attenuation ratio 1000:1 were used to measure the voltage across resistor one. Here all the output voltages of attenuators were voltages compared with the ground voltage. Resistor one was special, which could stand high voltage. The signals of measured voltages were digitalized, recorded and stored by Tektron digital oscilloscope. Three channels were used here with each channel generating a data file. The sampling rate was set to be 1.25 k per second throughout all the tests. In order to test the reliability and accuracy of the power supply, a sample was applied by gradually increasing voltage until discharge happened. The voltage signal extracted from the attenuator connected with the cathode was stable as shown in Fig 5.9 (a), even as the discharge happened as shown in Fig.6.9 (b). This showed the power supply had good reliability in providing with high voltage.
During the tests, samples were submerged in the silicone oil to prevent possible air discharge. In order to exclude any air bubble which might be attached to the sample surfaces, samples were manually vibrated in the silicone oil until no rising air bubbles were observed with the naked eyes. Voltage was gradually applied manually and recorded through three channels of the digital oscilloscope. Black paths, which connected the notch tip(s), and the opposite electrode, clearly appeared on the sample surfaces as shown in Fig.6.10. Those paths might reveal the traces of the breaking down behavior. Lightening sparkles were observed during the time when the samples were suffering the dielectric failure (see Fig 6.11). Typical voltage change on both anode side and cathode side was shown in Fig.6.12. The pulses on the cathode side were not necessarily matched by pulses from the anode side. The voltage just before a discharging was defined as the breakdown voltage here. For intensive discharging cases, silicone oil was blackened by possible products of chemical reaction.

6.4 Finite element analysis

Commercial codes, Ansys, were used to calculate the intensity factor of electrical strength $K_e$ for various crack lengths. 8-node plane element for electrostatic solids was used. In order to obtain accurate numerical results, the elements near the crack tip were refined step by step till their size was about one-millionth of the characteristic length of the samples (as shown in Fig 6.13). The intensity factors were evaluated by
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\[ K_e = \sqrt{2\pi x E_x} \quad 0 < x < \delta \quad (1) \]

Where \( E_x \) denotes the electric strength in the \( x \) direction, i.e., the direction of the conductive notch. \( \delta \) denotes a sufficiently small length. An example was given in Fig. 6.14.

Here we directly gave relationship between \( K_e \) and crack length, obtained by fitting the finite element analysis (FEA) results, as:

\[
K_e = \frac{V}{\sqrt{w}} f(L/w)
\]

\[
= \frac{V}{\sqrt{w}} \left( -232.3645 + 874.6168 \left( \frac{L}{w} \right) - 1093.0482 \left( \frac{L}{w} \right)^2 + 458.4124 \left( \frac{L}{w} \right)^3 \right) \quad 2 \text{ (a)}
\]

*with* \( 15 \text{ mm} < L < 19 \text{ mm} \)

for the CT samples and

\[
K_e = \frac{V}{\sqrt{w}} f(L/w)
\]

\[
= \frac{V}{\sqrt{w}} \left( -0.17334 + 19.75184 \left( \frac{L}{w} \right) - 109.28477 \left( \frac{L}{w} \right)^2 + 294.15642 \left( \frac{L}{w} \right)^3 \right) \quad 2 \text{ (b)}
\]

*with* \( 0.5 \text{ mm} < L < 5.0 \text{ mm} \)

for the double notched samples.
6.5 Results and Discussion

With experiment results and Eq.2, the electrical fracture toughnesses for both CT samples and double notched samples were obtained. The relationships between electrical fracture toughnesses $K_e$ and the notch lengths were plotted in Fig. 6.15 for the CT samples and in Fig 6.16 for the double notched samples. Notch lengths for CT samples and double notched samples were within a range from 16.2 mm to 17.7 mm, and a range from 3.8 mm to 4.8 mm, respectively. The mean value of the electrical fracture toughnesses for the CT samples was $317.7 \pm 37.1 \text{kV} \sqrt{m}$ with a relative error of ±11.7%. For the double notched samples, the mean value was $283.5 \pm 32.5 \text{kV} \sqrt{m}$ with a relative error of ±11.5%. $t$-test showed that the two groups of data were significantly different and could not be statistically treated as one group. This means the electrical fracture toughnesses for CT samples are higher than those for the double notched samples. For both cases, the mean values of the electrical fracture toughnesses of PVC were almost double the value of that for poled PZT 8 [22].

The test results indicated that the electrical fracture toughnesses were different for the same material PVC but different sample geometries. This was in accordant with the series of results of the experimental study conducted on PE material [6-9]. Charge density profiles across the PE thin films were measured using the PEA technique [6-9] and showed both positive charge carriers and negative charge
carriers were injected into the films, and the positive charge carriers had much higher mobility. After the films were electrically stressed for some time, the charge density profiles were unsymmetrical due to the quicker migration of the positive charge carriers. The electrical fracture tests on bulk PE material also gave accordant results. Using the pin-plate test method [17], it was found the tree initiation voltage and breakdown voltage were, respectively, 37 kV and 39 kV when the pin was positive in polarity. Both values were significantly lower than those when the pin was negative in polarity that were 54 kV and 100 kV, respectively. The apparent differences were explained by considering the behavior of charge carriers under the influence of electric stresses. When the pin was negative, electrons injected from the pin tip moved in a divergent deceleration field and quickly became trapped. The trapped electrons would form a homo-space charge, tending to reduce the effective field at the injecting contact. When the pin was positive, holes injected from the pin tip also moved in a deceleration field, but they traveled longer distance before being trapped because holes had higher mobility in the valence band. Trapped electron space charge was closer to the injecting contact. The efficiency of suppressing further hole injecting was not as good as when the electrode was negatively biased.

Introducing cut-through notches simplified the case to be two dimensional. For the CT samples, which were positively polarized here, holes were the emitted charge carriers. Compared with electrons, holes have much higher mobility and migrate longer distance, the suppressing effects are less significant. As a consequence, the DB voltage was relatively higher. For the double notched samples, both electrons and holes are emitted charge carriers and made contributions in the electrical failures.
Different dominant emitted charge carriers have different behavior under the influence of electric field and result in different DB voltage. Therefore, electrical failures are not only related to the material itself, but also the geometry of samples.

6.6 Conclusions

To investigate the electrical failures of polymeric vinyl chloride (PVC), two kinds of samples, CT samples and double notched samples, were used to conduct tests and the electrical fracture toughnesses were obtained. Electrical fracture toughnesses obtained from the CT samples were higher than those from the double notched samples. Experiment results showed that for the same PVC, the electrical fracture toughnesses were related to the sample geometry and how the notch tips were electrically biased.
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References:

6. Teruyoshi Mizutani, Space charge measurement and space charge in polyethylene, IEEE Transactions on Dielectrics and Electrical Insulation Vol. 1 No.5, October 1994
7. T. Mizutani, H. Semi, K. Kaneko, Space charge behavior in Low density polyethylene, IEEE Transactions on Dielectrics and Electrical Insulation, Vol. 7 No. 4, August 2000
8. N. Hozumi, T. Takeda, H. Suzuki and T. Okamoto, Space charge behavior in XLPE cable insulation under 0.2-1.3 MV/cm dc fields. IEEE Transactions on Dielectrics and Electrical Insulation Vol. 5 No. 1, February 1998


Fig. 6.1 Energy states before and after contact

($E_{Fm}$ Fermi energy of metal, $\phi_m$ the working function of metal, $\chi$ the electron affinity, $E_F$ Fermi energy of semiconductor, $\phi$ the working function of semiconductor, $E_v$ the upper energy level of valence band)

Fig.6.2 Band energy under biased voltage
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Fig. 6.3 Schematic of energy state density

Fig. 6.4 Comparison of energy states between ordered and disordered polymers

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Fig. 6.5 Schematic energy states in polymers

Fig. 6.6 Deep and shallow traps
Fig. 6.7 Schematic of polymer microstructures

Fig 6.8.a Pre-double notched samples

Fig.6.8 b Pre-notched CT samples
Fig. 6.9 a Power supply’s output voltage

Fig. 6.9 b Voltage change during dischargeing
Fig. 6.10 a  Electrically Failed CT sample

Fig. 6.10 b  Electrically failed double notched sample
Fig. 6.11 a, b, c Snapshots of the failure process
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![Graph showing anode and cathode voltages over time.](image)

**Fig. 6.12** Cathode and anode voltages

![FEA meshed object](image)

**Fig. 6.13** The FEA meshed object
Fig. 6.14. $E_x \sqrt{2 \pi s}$ vs the distance from crack tip

Fig. 6.15 Electrical fracture toughnesses for CT samples
Fig. 6.16 Electrical fracture toughnesses for pre-double notched samples
Chapter Seven

Summary and Future Works

The study aims at understanding the failure behavior of dielectric materials under mechanical/electrical loading, and establishing failure criteria which are important for reliability design in engineering and industries where dielectric materials like ceramics and polymers are involved. To achieve these objectives, the charge free zone (CFZ) model, which explained well for the failure behavior of depoled piezoelectric ceramics PZT 4, was extended to piezoelectric ceramics. The failure criterion was established for poled piezoelectric ceramics based on the CFZ model. Fracture tests on the piezoelectric ceramics PZT 8 were conducted to verify the theoretical prediction from the CFZ model. To refine the CFZ model for the poled piezoelectric ceramics, the concept of secant piezoelectric constant was introduced. In addition, electrical failures of polymeric material poly vinyl chloride (PVC) under different electric loading conditions were also investigated.

7.1 Charge free zone (CFZ) model for piezoelectric ceramics

Charge carriers injected into and then trapped in the dielectric materials from electrode(s) have great influence on the fracture behavior. For conductive crack problems, the trapped charges form a homo-space charge zone and partially shield the crack tip from applied electric field. To include this physical phenomenon and to evaluate charge carriers’ influence, the charge free zone (CFZ) model, which was proposed by Zhang et
to understand the fracture behavior of doped PZT 4 under the mechanical and/or electrical loading, was extended for the conductive crack problems of piezoelectric ceramics. The CFZ model yields an explicit failure criterion to predict the failure behavior of conductive cracks in piezoelectric ceramics under electrical and/or mechanical loading. The failure formula can be mathematically expressed in the form of an elliptical equation \( x^2 + \eta xy + y^2 = 1 \), where \( x \) and \( y \) denote the normalized mechanical fracture toughness and normalized electrical fracture toughness, respectively. For doped piezoelectric ceramics \( \eta \) disappears, the failure formula takes form of \( x^2 + y^2 = 1 \).

7.2 Fracture tests on piezoelectric ceramics PZT 8

Compact tensile (CT) test method was adopted to investigate the fracture behavior of piezoelectric ceramics PZT 8 under the mechanical and/or electric loading. Tests were conducted under electrically positive loading and electrically negative loading. Finite element method (FEM) was used to calculate the relationship between the mechanical/electric intensity factors and the crack length. With the measured critical load and/or critical voltages at the onset of failure, the mechanical fracture toughnesses and the electric fracture toughnesses were obtained. The critical energy release rates were obtained from the fracture toughnesses. The energy release rates for the conductive cracks were, respectively, \( 164.3 \pm 29.9 \, N/m \) and \( 181.6 \pm 25.94 \, N/m \) under the purely positive and negative electric fields. The values of energy release rates for both purely positive and negative cases were much higher than the mechanical energy release rate. The reason for the big differences in the toughness may be due to the fact that
piezoelectric ceramics are mechanically brittle and electrically ductile. The normalized mechanical/electrical fracture toughnesses under combined mechanical and electrical loading were analyzed. The results indicated that the coupling term in the elliptical equation was -0.16 under negative electrical loading and 0.52 for under positive electrical loading. The experiment results verified the explicit failure criteria derived from the CFZ model.

The fractographs of the poled PZT 8 ceramics samples under various combinations of electrical and mechanical loading were examined. Black areas, which initiated from the crack fronts, were observed on the fractured surfaces. The observations showed that the large electrical load led to a large black areas.

7.3 Secant piezoelectric constant

The usually adopted piezoelectric constants are those measured at low electric field (voltage). In our tests, samples were electrically broken down at high electric field (voltage). Butterfly curves of piezoelectric ceramics show an apparent nonlinear relationship between strain (deformation) and electric field (voltage) when the electric field is high. In order to take those factors into account, the concept of secant piezoelectric constant was introduced to refine the CFZ model. The refined CFZ model was verified by experimental results of poled PZT 8. The normalized experimental results for poled PZT 4 were also in accordant with the theoretical prediction. Therefore, once again, the refined CFZ model was proved to be a solid and robust tool to understand and predict the failure of poled/depoled piezoelectric ceramics under mechanical/electrical loading.
Chapter 7  Summary and Future Works

7.4 Electric breakdown in polymeric vinyl chloride (PVC)

Polymers are used widely in high power engineering and the electrical failure study of polymers has attracted great attention of researchers. In the present work the concept of fracture mechanics was extended to study the failure behavior of a polymeric material, poly vinyl chloride (PVC). Two groups of samples, pre-notched compact tensile (CT) samples and double notched samples, were used to conduct the electrical failure tests. Experiment results showed that the electrical fracture toughnessees obtained from CT samples were higher than those from the double notched samples. And the differences in the electrical fracture toughnessees were well explained by the different behavior of charge carriers under the influence of electric field.

7.5 Future works

To better understand the electrical fracture failure of dielectric materials, observations and measurements of charge carrier behavior in dielectric materials are important and necessary. In addition, the local characters in dielectric materials may be not evenly distributed at the micro-level. The interaction between charge carriers and local characters of dielectric materials needs to be investigated.