RICE UNIVERSITY

Fracture Toughening of Ferroelectric Ceramics under Electro-Mechanical Loading

by

Jianxin Wang

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE

Doctor of Philosophy

APPROVED, THESIS COMMITTEE:

Chad M. Landis, Associate Professor, Chair
Mechanical Engineering & Materials Science

Michael M. Carroll, Professor
Mechanical Engineering & Materials Science

Satish Nagarajaiah, Professor
Civil & Environmental Engineering

HOUSTON, TEXAS

JULY 2006
INFORMATION TO USERS

The quality of this reproduction is dependent upon the quality of the copy submitted. Broken or indistinct print, colored or poor quality illustrations and photographs, print bleed-through, substandard margins, and improper alignment can adversely affect reproduction.

In the unlikely event that the author did not send a complete manuscript and there are missing pages, these will be noted. Also, if unauthorized copyright material had to be removed, a note will indicate the deletion.

UMI

UMI Microform 3256761
Copyright 2007 by ProQuest Information and Learning Company.
All rights reserved. This microform edition is protected against unauthorized copying under Title 17, United States Code.

ProQuest Information and Learning Company
300 North Zeeb Road
P.O. Box 1346
Ann Arbor, MI 48106-1346
ABSTRACT

Fracture Toughening of Ferroelectric Ceramics under Electro-Mechanical Loading

by

Jianxin Wang

In this dissertation, the fracture toughening behavior of ferroelectric materials under different electromechanical loading conditions is predicted and compared to available experimental observations. A multi-axial, electromechanically coupled, incremental phenomenological constitutive model for ferroelectric ceramics is developed first. The constitutive model is then implemented within the finite element method to study the effects of electric field on the Mode I steady crack growth under plane stress and plane strain conditions.

Toughening behaviors of electrically permeable cracks are simulated on both initially unpoled and poled materials with electric field applied in-plane or parallel to the crack front. The finite element results give detailed electromechanical fields, remanent strain and remanent polarization distributions, domain switching zone shapes and sizes, and the crack tip energy release rate. It is shown that the toughening is related to the size of the concentrated switching zone that is confined to a small region around the crack tip. The model predicts a range of phenomena that indicate that the toughening is dependent on both the level of electric filed applied and on the polarization state. In addition to the
effects of electric field, the effects of the plane-stress constraint and transverse stress are also established in the out-of-plane poled cases.

In a similar manner to the electrically permeable cracks, the crack growth simulations for electrically conducting crack face boundary conditions are also performed. The results predict the toughening variations under combined electromechanical loadings for poled or unpoled materials. The electromechanical fields from the finite element results are used to determine the stress and electric field intensity factors around the crack tip.

The favorable comparison of the present model to the experimental observations suggest that ferroelectric switching behavior is more accurately modeled with an incremental plasticity formulation, rather than as an unstable phase transformation. The nonlinear studies of this dissertation not only explain most available experimental phenomena but also enhance the understanding of the nature of fracture in ferroelectric ceramics.
ACKNOWLEDGEMENT

My first sincere appreciation is presented to my advisor, Dr. Chad M. Landis. Throughout my doctoral study, Dr. Landis provided invaluable help and suggestions, from hands on teaching to the guidance of my independent research. However, he serves not only an academic mentor but also a life model to me. I am greatly indebted for his care of my family and me.

Great thanks to the committee members, Dr. Michael M. Carroll and Dr. Satish Nagarajaiah, for their time and advice on this topic. They took invaluable time in discussion with me and gave many suggestions for the final shape of this thesis.

During the writing of the thesis, Dr. Jan Hewitt with the Cain Project gave me a lot of help. Thank you Dr. Hewitt! It’s great to work with my colleagues, Dr. Yu Su, Jianshun Sheng and Wenyuan Li. I want to thank them for the many useful discussions and the great times with them.

Finally, I owe many thanks to my family. Without my beautiful wife Xiuling’s support and caring, I would never be able to go so far. My sons, Kevin and the unborn one, are my great sources of energy and power!
# Table of Contents

Abstract .............................................................................................................. I

Acknowledgements .......................................................................................... IV

Table of Contents ............................................................................................ V

List of Tables ................................................................................................ IX

List of Figures ................................................................................................ X

1. Introduction ................................................................................................. 1
   1.1 Features of ferroelectric materials ......................................................... 2
   1.2 Current studies on ferroelectric materials ............................................. 10
      1.2.1 Constitutive modeling ..................................................................... 10
      1.2.2 Fracture studies of ferroelectric ceramics ....................................... 12
   1.3 Plan of the thesis ..................................................................................... 14

2. A phenomenological constitutive model for ferroelectrics .................... 15
   2.1 Introduction .......................................................................................... 15
   2.2 Governing equations ........................................................................... 16
   2.3 Formulation for the incremental constitutive model ......................... 18
   2.4 Results ................................................................................................. 24
      2.4.1 Pure electrical loading ................................................................. 24
      2.4.2 Pure mechanical loading ............................................................. 28
      2.4.3 Combined electromechanical loadings ....................................... 31
      2.4.4 Multi-axial polarization rotation tests ....................................... 36
   2.5 Summary ............................................................................................ 38
3. The fracture toughness of ferroelectric ceramics with an electric field applied parallel to the crack front ............................ 39

3.1 Introduction ................................................................. 39

3.2 The fracture model and finite element formulation ....................... 41

3.2.1 The loading process .................................................. 42

3.2.2 Small scale switching ................................................. 45

3.2.3 Finite element formulation ......................................... 49

3.3 Results ............................................................................ 50

3.3.1 Switching zones ....................................................... 52

3.3.2 The effect of initial polarization and electric field on toughening . 55

3.3.3 Plane strain versus plane stress effects ............................... 60

3.3.4 The T-stress effect ...................................................... 63

3.4 Discussion ....................................................................... 64

3.5 Summary ........................................................................... 66

4. Effects of in-plane electric fields on the toughening behavior of ferroelectric ceramics ................................................................. 67

4.1 Introduction ....................................................................... 67

4.2 The fracture model and finite element formulation ....................... 70

4.2.1 The loading process .................................................. 70

4.2.2 Boundary conditions .................................................. 73

4.2.3 Finite element formulation ......................................... 74

4.3 Results ............................................................................ 77

4.3.1 Switching zones ....................................................... 77
4.3.2 The effect of initial electrical polarization on toughening ............ 80

4.3.3 Effect of applied electric field on toughening: the perpendicular poling case ................................................................. 83

4.3.4 Effect of applied electric field on toughening: the parallel poling case .................................................................................. 87

4.4 Discussion ................................................................................. 91

4.5 Summary ................................................................................... 93

5. Fracture toughening behavior for conductive crack growth in ferroelectric ceramics ......................................................................... 95

5.1 Introduction .............................................................................. 95

5.2 Conductive boundary conditions .................................................. 98

5.3 Results .................................................................................... 101

5.3.1 Switching zones .................................................................... 102

5.3.2 The K fields under combined loading ..................................... 104

5.3.3 Purely mechanical loading and purely electrical loading ............ 108

5.3.4 Toughening behavior under mixed mode loadings .................. 110

5.4 Discussion .............................................................................. 112

5.5 Summary ................................................................................... 113

6. Conclusions and future work ........................................................ 114

6.1 Conclusions .............................................................................. 114

6.2 Suggestions for future research .................................................. 116

Appendix A Solving the incremental constitutive model using the Newton-Raphson method ................................................................ 118
Appendix B Fracture toughening during steady state crack growth ......................................................... 121

Appendix C Detailed finite element formulation ................................................................. 124

Appendix D Asymptotic fields for the permeable and conducting cracks ................................................................. 127

References ........................................................................................................................................ 131
List of Tables

Table 5.1 The ratio of energy release rate under purely electrical loading $G^e$ and energy release rate under purely mechanical loading $G^m$ from the results of finite element simulation and recent tests

.................................................. .................................................. 110
List of Figures

Figure 1.1: Change of unit cell of BaTiO$_3$ when cooling from above the Curie temperature under load free condition. The vector of spontaneous polarization $P_3$ is oriented in the direction of the displaced titanium ion  

Figure 1.2: Schematic illustration of the pyroelectric effect  

Figure 1.3: Plane view of a crystal aggregate with domains as subregions of equal spontaneous polarization after cooling below the Curie temperature (Adapted from Kamlah, 2001)  

Figure 1.4: Schematic diagram of the 90 or 180 degree switching of a unit cell under electric field loading  

Figure 1.5: A typical schematic diagram of ferroelectric hysteresis loop. Here $P_s$ – spontaneous polarization; $P_r$ – remanent polarization; $E_c$ – coercive field  

Figure 1.6: Poling of the aggregate shown in Figure 1.2. After unloading, the domain structure remains in the switched state, now possessing a resultant residual macroscopic polarization and, thus, exhibiting a macroscopic piezoelectric effect  

Figure 1.7: Schematic diagram of possible polar reorientation under uniaxial compression and tension (Adapted from Kamlah, 2001)  

Figure 2.1: Prediction of the electric displacement versus electric field hysteresis loop under uniaxial electric field loading at zero stress. The domain structures at states A-G are for the purpose of explanation.
Figure 2.2: Prediction of the strain versus electric field butterfly loop corresponding to the hysteresis loop under uniaxial electric loading at zero stress. States A-G here are the same as those in Figure 2.1 .......................................................... 27

Figure 2.3: Simulation of ferroelastic behavior: uniaxial stress versus longitudinal strain hysteresis loop at zero electric field ....................................................... 30

Figure 2.4: Stress versus electric displacement during the simulation of mechanical depolarization of poled specimen .............................................................. 31

Figure 2.5: The counterpart of Figure 2.4; stress versus strain during the simulation of mechanical depolarization of poled specimen ................................. 33

Figure 2.6: Predictions of polarization versus electric field and strain versus electric field hysteresis loops under different constant preloaded compressive stress. The initial states in (A), (B), and (C) correspond to the state of A, B, and C in Figure 2.4 ................................................................................. 35

Figure 2.7: Polarization rotation tests obtained from the constitutive law. Left: Change of the electric displacement versus electric field. Right: Change of strain versus electric field .................................................................................. 37

Figure 3.1: A schematic of the out-of-plane electrical and in-plane mechanical configuration to be modeled in this work. For any given sample, the electric field and remanent polarization are aligned in x-direction. The in-plane mechanical loading is simply indicative of the Mode I symmetry to be modeled in this work and should not be interpreted literally ............ 40

Figure 3.2: The uniaxial electromechanical behavior of the model material with three levels of the poling field $E_{x}^{p}$ leading to different partially poled states. (a)
The electric field versus electrical displacement hysteresis loops. (b) The
electric field versus strain butterfly loop. (c) The stress versus electrical
displacement depolarization loop. (d) The stress versus strain loop during
depolarization. Notice that the intermediate lines in (a) and (b) represent the
response during the unloading of electric field, and those in (c) and (d)
represent the depolarization behavior from a partially poled state .......

Figure 3.3: Switching zone sizes and shapes for initially unpoled (a,b) and initially
poled (c) cases. Figures (d) and (e) can be interpreted as for either initially
poled or initially unpoled samples. The outermost curved contours on these
plots delineate the location of the active switching boundary. The inner
solid contours give the location within the active switching zones where the
effective remanent strain change achieves the characteristic elastic level of
$$\Delta \varepsilon' = \sqrt{2 \Delta \varepsilon_y' \Delta \varepsilon_z' / 3} = \sigma_0 / E.$$ The inner dashed contours give the location
within the active switching zones where the change of remanent polarization
achieves the characteristic linear dielectric level $$|\Delta P_3'| = \kappa E_0.$$ Notice that the
spatial coordinates are normalized by $$R_0,$$ which is the characteristic
switching zone size when the applied energy release rate reaches $$G_0,$$ i.e.
$$R_0 = G_0 E'/3\pi\sigma_0^2.$$ Therefore, if $$G_0$$ is the same for all cases, then the spatial
coordinate normalizations for each plot are identical ......................

Figure 3.4: The normalized toughness enhancement $$G_a/G_0$$ versus the applied out-of-
plane electric field for a range of initial poling states for plane-strain
conditions. The bold solid line and bold dashed line on the main plot
correspond to the solid and dashed lines on the outer hysteresis and butterfly

XII
loops depicted on the inserts in the upper left and right corners. The thin
tables represent the toughening behavior of unpoled or partially poled
materials. Points A and B are highlighted to indicate the relationships
between the electromechanical constitutive response and the fracture
toughness predictions ................................................. 56

Figure 3.5: A comparison of the effects of plane-strain versus plane-stress out-of-plane
mechanical constraint on the toughness enhancement of partially poled
materials. The normalized toughness enhancement $G_s/G_0$ is plotted versus
the initial poling field $E_0^p/E_0$. For all cases the initial poling field is
removed and no subsequent electric field is applied ..................... 61

Figure 3.6: The effects of the transverse stress $T_{11}$ on the toughness enhancement under
plane strain conditions. The normalized toughness enhancement $G_s/G_0$ is
plotted versus the applied out-of-plane electric field with or without $T$-stress
for (a) initially unpoled and (b) initially poled with $P_3^r/P_0 = 0.88$ .... 63

Figure 4.1: A schematic of the in-plane electrical and in-plane mechanical loading
configuration to be modeled in this work. For any given sample, the electric
field and remanent polarization are aligned in $x_1$ or $x_2$ direction. The in-
plane mechanical loading is simply indicative of the Mode I symmetry to be
modeled in this work and should not be interpreted literally ............. 67

Figure 4.2: The uniaxial electromechanical behavior of the model material with three
levels of the poling field, leading to different partially poled states. (a) The
electric field versus electrical displacement hysteresis loops. (b) The
electric field versus strain butterfly loop. (c) The stress versus electrical
displacement depolarization loop. (d) The stress versus strain loop during depolarization. Notice that the intermediate lines in (a) and (b) represent the response during the unloading of electric field, and those in (c) and (d) represent the depolarization behavior from a partially poled state ........ 71

Figure 4.3: Switching zone sizes and shapes for initially unpoled (a) and initially poled (b, c, d, e) cases. The outermost curved contours on these plots delineate the location of the active switching boundary. The inner solid contours give the location within the active switching zones where the effective remanent strain change achieves the characteristic elastic level of \( \Delta \varepsilon^r = \sqrt{2 \Delta \varepsilon_i^r \Delta \varepsilon_0^r / 3} = \sigma_0 / E \). The inner dashed contours give the location within the active switching zones where the change of remanent polarization achieves the characteristic linear dielectric level \( |\Delta P'| = \kappa E_0 \). Notice that the spatial coordinates are normalized by \( R_0 \), which is the characteristic switching zone size when the applied energy release rate reaches \( G_0 \), i.e. \( R_0 = G_0 E'/3\pi\sigma_0^2 \). Therefore, if \( G_0 \) is the same for all cases, then the spatial coordinate normalizations for each plot are identical .................... 78

Figure 4.4: A comparison of the effects of poling direction on the toughness enhancement of partially poled materials. The normalized toughness enhancement \( G_{\alpha i} / G_0 \) is plotted versus the initial remanent polarization state \( P_{i o} / P_0, i = 1, 2, 3 \) ................................................................. 82

Figure 4.5: Normalized toughness enhancement \( G_{\alpha i} / G_0 \) versus the applied electric field \( E_2 \) for a range of initial poling states for plane-strain conditions. The
colored lines on the main plot are related to the corresponding colored lines on the hysteresis and butterfly loops depicted on the inserts in the upper left and right corners. Points A and B are highlighted to indicate the relationships between the electromechanical constitutive response and the fracture toughness predictions.

Figure 4.6: The normalized toughness enhancement $G_a/G_0$ versus the applied electric field $E_i$ for a range of initial poling states for plane-strain conditions. The colored lines on the main plot correspond to the corresponding colored lines on the hysteresis and butterfly loops depicted on the inserts in the upper left and right corners. Points A and B are highlighted to indicate the relationships between the electromechanical constitutive response and the fracture toughness predictions.

Figure 5.1: A schematic illustration of a conductive crack. The plus and minus signs here represent electric charges. $E_i$ is the applied electric field.

Figure 5.2: A schematic of the electromechanical configuration with a conductive crack to be modeled in this work. For any given sample, the electric field and remanent polarization are aligned in $x_i$ direction. The in-plane electromechanical loading is indicative of the Mode I symmetry to be modeled in this work and should not be interpreted literally.

Figure 5.3: Switching zone sizes and shapes for initially unpoled (a, b) and initially poled (c, d) cases. The outermost curved contours on these plots delineate the location of the active switching boundary. The inner solid contours give the location within the active switching zones where
Figure 5.7: Normalized toughness enhancement $G_a/G_0$ versus the ratio of $K_a/K_0$ for a range of initial poling states for plane-strain conditions. The initial poling level $P_0$ under mechanical loading only.

Figure 5.6: Normalized toughness enhancement $G_a/G_0$ versus the ratio of initial $K_a/K_0$.

Figure 5.5: The normalized crack tip intensity factor $K_j$ versus the ratio of initial crack tip intensity factor $K'_j$ during combined loadings: (a) $K^\text{tip}_j/K_0$ versus $K'_j/K_0$; (b) $K^\text{tip}_j/K_0$ versus $K'_j/K_0$.

Figure 5.4: The normalized applied intensity factor during combined loadings: $K_j/K_0$.

\[ \Delta \varepsilon = \frac{\sqrt{3}}{2} (\Delta \varepsilon_0, \Delta \varepsilon_0) = \sigma_0 E. \]  

The inner dashed contours give the location within the active switching zones where $\Delta \varepsilon^p = \Delta \varepsilon_0$. Notice that the spatial coordinates are normalized by $\Delta \varepsilon_0$, which is the characteristic switching zone size when the applied energy release rate reaches $G^s_0$, i.e., $R_j = G^s_0/E/\Delta \varepsilon_0^2$. Therefore, if $G^s_0$ is the same for all cases, then the spatial coordinate normalizations for each plot are identical.
Chapter 1 Introduction

Recently, ferroelectric materials have attracted much attention because of their extraordinary dielectric properties and electromechanical performance. Their applications in smart systems have promoted ferroelectric ceramics to become a technologically important class of materials. The primary applications (Safari et al.) of ferroelectric ceramics have been for capacitor applications, ferroelectric thin films for non-volatile memories, piezoelectric materials for medical ultrasound imaging and actuators, and electro-optic materials for data storage and displays. Along with the discovery and new synthesis techniques (Safari et al.) for ferroelectric materials have come various experimental investigations into their properties and fracture behaviors (Zhang et al., 2001; Chen and Lu, 2002). In order to gain an understanding of their fundamental characteristics, theoretical modeling is equally essential. Accurate modeling tools are needed for the reliable design and optimized performance of ferroelectric devices. However, theoretical research on these materials can be quite complicated due to the complex crystal structure of ferroelectric ceramics, the electromechanical coupling effects and the non-linear behavior associated with domain switching. The present study provides a phenomenological model for the constitutive behavior of ferroelectric materials and investigates their fracture toughening behavior under different electromechanical loading conditions.

In order to better understand the different behaviors and theoretical results, some basic features of ferroelectric materials will be introduced in the next section. A brief
review of the current studies on constitutive modeling and fracture behavior of ferroelectric ceramics is then given. Finally, the outline of the thesis is listed.

1.1 Features of ferroelectric materials

Ferroelectricity is a phenomenon first discovered in 1921 by Valasek on a single crystal Rochelle salt. A huge leap in the research on ferroelectric materials came in the 1950's, leading to the widespread use of barium titanate (BaTiO₃) based ceramics in capacitor applications and piezoelectric transducer devices. Since then, many other ferroelectric ceramics including lead titanate (PbTiO₃), lead zirconate titanate (PZT), lead lanthanum zirconate titanate (PLZT), and relaxor ferroelectrics like lead magnesium niobate (PMN) have been developed and utilized for a variety of applications (Buchanan, 1986). Ferroelectricity is defined as the presence in a crystal of a spontaneous electric moment which can be changed in its orientation between two or more distinct crystallographic directions by applying an external electric field. Materials presenting the feature of "ferroelectricity" are called "ferroelectric" or ferroelectric materials. The name "ferro" refers to certain analogies between ferroelectric phenomenon and ferromagnetism, though it is somewhat misleading as it has no connection with iron (ferrum) at all. The similarity is phenomenological: ferromagnetic materials exhibit a spontaneous electric magnetization and hysteresis effects in the relationship between magnetization and magnetic field; whereas ferroelectric crystals show a spontaneous electric polarization and hysteresis effects in the relation between displacement and electric field. Ferroelectricity has also been called Seignette electricity, as Seignette or
Rochelle Salt (RS) was the first material found to show ferroelectric properties such as a spontaneous polarization on cooling below the Curie point, ferroelectric domains and a ferroelectric hysteresis loop (Valasek, 1921).

Ferroelectric materials also exhibit the piezoelectric effects. Piezoelectricity is the ability of certain crystalline materials to develop an electrical charge proportional to a mechanical stress. The application of pressure to a piezoelectric crystal causes a charge to flow in a certain direction. If the pressure is replaced by a tension, the charge will flow in the opposite direction. Piezoelectric materials also show a converse effect, where a geometric strain (deformation) is produced by the application of a voltage. If an electric field is applied, the crystal will be stretched; if the electric field is reversed, it will be shortened. One has to keep in mind that the piezoelectric effect is a linear effect and only those piezoelectric materials which show spontaneous polarization are ferroelectrics.

The spontaneous polarization is given by the value of the dipole moment per unit volume or by the value of the charge per unit area on the surface perpendicular to the axis of spontaneous polarization. To better understand the concept of spontaneous polarization, a unit cell of typical ferroelectric material barium titanate (BaTiO₃) as shown in Figure 1.1 is used as an illustration. To understand the electromechanical properties of a material, the knowledge of the position of the centers of the charges relative to each other in the unit cell is crucial. A material is called polarizable, if the centers of positive and negative ions in the unit cell can be shifted with respect to each other by an external load, leading to a load induced dipole of the unit cell. If the centers of positive and negative charges are at different positions within the unit cell in the absence of any load giving rise to a permanent dipole, we say that the cell possesses
spontaneous polarization.

Figure 1.1: Change of unit cell of BaTiO$_3$ when cooling from above the Curie temperature under load free condition. The vector of spontaneous polarization $P_s$ is oriented in the direction of the displaced titanium ion.

Figure 1.2: Schematic illustration of the pyroelectric effect.
The value of spontaneous polarization depends on the temperature, a phenomenon called the pyroelectric effect. Pyroelectric crystals show a spontaneous polarization in a certain temperature range. If the magnitude and direction of spontaneous polarization can be reversed by an external electric field, then such crystals are said to show ferroelectric behavior. All single crystals and successfully poled ceramics which show ferroelectric behavior are pyroelectric, but not vice versa. The polarization suddenly falls to zero on heating the crystal above the Curie point as shown in Figure 1.2.

All ferroelectric materials have a transition temperature called the Curie point $T_c$. Consider, for example, BaTiO$_3$ in Figure 1.1. Above the material-dependent Curie temperature $T_c$, the crystal lattice is of a so-called perovskite structure. The unit cell is in cubic shape with side length $a_0$ and a four times positively charged ion in the center. The centers of positive and negative charges coincide and the material exhibits no spontaneous polarization. This cubic state is called the paraelectric phase and no piezoelectricity will be observed. If the temperature is below $T_c$, the unit cell is displaced parallel to one of the edges and deformed to a tetragonal shape. In this ferroelectric phase, the centers of positive and negative charges no longer overlap, the unit cell possesses a spontaneous polarization and, consequently, piezoelectric properties. As we see, on decreasing the temperature through the Curie point, a ferroelectric crystal undergoes a phase transition from a paraelectric phase to a ferroelectric phase. The unit cell in crystals will change from high symmetry to lower symmetry. For crystals such as BaTiO$_3$ and PZT, the crystal structure is in cubic form above the Curie temperature; it will evolve to tetragonal, orthorhombic, and rhombohedral structures when the...
temperature continues to drop below the transition point.

Ferroelectric crystals possess regions with uniform polarization called ferroelectric domains. Within a domain, all the electric dipoles are aligned in the same direction. Usually typical materials like BaTiO$_3$ and PZT are not produced as ideal single crystals. Instead, those ceramics have a polycrystalline structure with grains of differently oriented crystal lattices. There may be many domains in a grain/crystal separated by interfaces called domain walls as shown in Figure 1.3.

![Figure 1.3](image)

Figure 1.3: Plane view of a crystal aggregate with domains as subregions of equal spontaneous polarization after cooling below the Curie temperature (Adapted from Kamlah, 2001).

A strong applied electric field can lead to the rotation or reversal of the polarization within a domain, known as 90 degree or 180 degree domain switching, as shown in Figure 1.4. The polarization reversal can be observed by measuring the ferroelectric hysteresis. Figure 1.5 shows the typical schematic diagram of ferroelectric hysteresis loop. The
hysteresis behavior of ferroelectric materials is the basis for applications in smart systems and is the primary material behavior of interest in this thesis. As the electric field strength is increased, the poling process arranges the formerly random polarization orientation state of the unit cells towards the direction of the poling field. The favorable oriented domains grow giving rise to a rapid increase in the polarization (OB). At very high field levels, the polarization reaches a saturation value ($P_{sat}$) due to the constraint of the crystal.

Figure 1.4: Schematic diagram of the 90 or 180 degree switching of a unit cell under electric field loading.
structure. The polarization does not fall to zero when the external field is removed. At zero external field, some of the domains remain aligned in the original field direction. This macroscopic average of the spontaneous polarization of the unit cells is called remanent polarization \( P_r \). The presence of remanent polarization when decreasing the electric field from a high value to zero is illustrated in Figure 1.6. The crystal cannot be completely depolarized until a field of magnitude \( OF \) is applied in the negative direction. This external field needed to reduce the polarization to zero is called the coercive field strength \( E_c \). If the field is increased to a higher negative value, the direction of polarization flips and a hysteresis loop is obtained. The value of the spontaneous polarization \( P_s \) (OE) is obtained by extrapolating the curve onto the polarization axes (CE).
Figure 1.6: Poling of the aggregate shown in Figure 1.2. After unloading, the domain structure remains in the switched state, now possessing a resultant residual macroscopic polarization and, thus, exhibiting a macroscopic piezoelectric effect (Adapted from Kamlah, 2001).

Figure 1.7: Schematic diagram of possible polar reorientation under uniaxial compression and tension (Adapted from Kamlah, 2001).

As for the electrical loading, mechanical loading can also cause polar reorientation of unit cells. As shown in Figure 1.7 the dielectric dipole will switch to the direction perpendicular to the original poling axis when it is under a uniaxial compression along the
axis or uniaxial tension perpendicular to the axis. For a poled unit cell with tetragonal structure, there are four potential final orientations under uniaxial compression and two potential final orientations under uniaxial tension. Combination of mechanical and electric loading will also cause similar types of reorientation. Though such cases under combined loading are much more complicated, the switching mechanism can be used to explain many phenomena including the fracture toughening behavior that is presented in this thesis.

1.2 Current studies on ferroelectric materials

Ferroelectric ceramics have drawn much attention in recent years due to their high potential for applications in accuracy sensors, actuators, and memory storage. To predict the performance and reliability of ferroelectric devices, a number of problems require the resolution of the inhomogeneous electrical and mechanical fields in the material. Such problems range from the prediction of poling patterns around electrodes in ferroelectric actuator designs to the determination of switching zones near crack tips that govern failure in these materials. Since a reliable constitutive model is the basis for fracture simulations, a brief review of both constitutive modeling and fracture analysis of ferroelectric materials is given in the following.

1.2.1 Constitutive modeling

A number of micro-electromechanical methods exist that model the behavior of ferroelectric polycrystals as the averaged response of randomly oriented single crystal grains. In all cases, the constitutive response of the single crystals is intended to model
the domain switching mechanism that gives rise to remanent strain and polarization in these materials. Then, the polycrystalline averages range in sophistication from Taylor and Reuss-like models where interaction between crystals is neglected (Hwang et al., 1995; Lu et al., 1999), to self-consistent methods where grain to grain constraints are accounted for in an average sense (Hwang et al., 1998; Huber et al., 1999), to finite element simulations of many cubic single crystals where interactions are fully accounted for (Hwang and McMeeking, 1998, 1999).

Even in today's applications in microsystems technology, the dimensions of ferroelectric devices belong to an order of magnitude in which the constitutive behavior is characterized by the macroscopic polycrystalline properties. In structural mechanics analyses, such constitutive behavior can be represented numerically most efficiently by constitutive laws constructed by phenomenological methods directly on the macro-level. Such models are appropriate for tracing the behavior of a device back to some characteristic macroscopic material constants. For the phenomenological models developed during recent years, the starting point is a free energy function with carefully chosen internal variables. Usually the models assume the additive decomposition of polarization and strain into linear and remanent parts. The formulations of the evolution equations for the internal variables can be based on an update algorithm during the finite element iteration (Ghandi and Hagood, 1997), or based on an equivalent stress, including contributions of the stress tensor and the electric field vector (Fan et al., 1999), or derived from the formulation of the release of dielectric energy due to switching (Drescher et al., 2000), or obtained from a yield function by the normality rule and consistency condition (Cocks and McMeeking, 1999; Landis, 2002a).
For more details of recent developments in constitutive modeling of ferroelectrics, please refer to the review articles (Kamlah, 2001; Landis, 2004a). Although phenomenological modeling can be more efficient when doing the simulation, it cannot, a priori, predict certain features of behavior such as the saturation conditions for the remanent polarization and remanent strain. The micromechanical models can do such predictions but is time-consuming when simulating the polycrystal behavior. A micro-electro mechanically informed phenomenological model represents an accurate and usable solution. In this thesis, we extend the phenomenological constitutive framework of Landis (2002a) by implementing the results of possible remanent saturation states from a micro-mechanical model (Huber et al., 1999). The model is able to simulate several different experimental results.

1.2.2 Fracture studies of ferroelectric ceramics

A significant body of work has appeared in the last few decades on the fracture mechanics of ferroelectric materials. The techniques used to experimentally investigate fracture in these materials include Vickers indentations (Lynch, 1998; Wang and Singh, 1997), four point bend tests (Chen et al., 2001), the double torsion technique (McHenry and Koepeke, 1983) and compact tension tests (Kolleck et al., 2000; Lucato et al., 2002). Thorough reviews of the literature can be found in Zhang et al. (2001) and Chen and Lu (2002). It was shown that electrical boundary conditions, polarization, and electric field can all affect the R-curve behavior (Lucato et al., 2002).

In fracture mechanics, the concept of the energy release rate is extremely important because it provides a clear physical picture of the energetic of crack growth. Many
researchers, as for example, Suo et al. (1992), McMeeking (1999), Landis (2004c), and Landis (2004d), have specifically studied the energy release rate for cracks within the context of linear piezoelectricity. These studies show that the evaluation of the energy release rate can be significantly affected by the choice of the electrical boundary conditions on the crack faces. The basic electrical boundary condition can be classified as insulating or conducting. Among the different modeling schemes, three approaches for the insulating crack problem have received considerable attention: (1) the impermeable crack which sets the normal component of the electric displacement to zero at the upper and lower crack faces, (2) the permeable crack which requires the electric potential and the electric displacement normal to the crack surface to be continuous across the slit, and (3) the semi-permeable boundary condition which treats the crack gap as a small vacuum filled capacitor. Most of these models implement the mechanical traction free condition on the crack surface. However, Landis (2004d) implements energetically consistent boundary conditions and his derivation shows that an additional closing traction should be added to the semi-permeable boundary conditions. Landis also showed that the traction free permeable conditions are recovered if the electrical breakdown strength of the crack gap medium is small. In the current study, the reasonable approximation of permeable electrical condition and traction free condition are used.

Several researchers have attempted to explain the toughening behavior of ferroelectrics under the influence of electric field. These models use the concept of transformation toughening (e.g. Yang and Zhu, 1998) or are based on the balance of the energy supplied by the driving forces and that consumed by dissipation (Kreher, 2002). However, due to the lack of a convincing constitutive law, either the details of the
mechanical and electrical coupling behavior of the material are neglected or the calculation of the complicated electromechanical crack tip fields is avoided. In this study, our model amends these simplifications by applying a recently developed phenomenological constitutive law for ferroelectric switching within the finite element method to determine the details of the crack tip fields and rigorous calculations of the toughening due to domain switching during steady crack growth.

1.3 Plan of the thesis

In Chapter 2, the balance laws and constitutive equations for ferroelectric materials are given first. A plasticity-like phenomenological constitutive law using the flow rule and a switching surface is then developed. The flow potential functions apply the results from micro-mechanical calculations. Simulations of the material behavior are obtained and compared to the experiments. In the following Chapters, this constitutive model is implemented into a finite element code and the fracture behaviors of ferroelectric materials under different poling and loading conditions are studied. Chapter 3 is devoted to the fracture toughness of ferroelectric ceramics with electric field applied parallel to the crack front. Chapter 4 studies the effects of in-plane electric field on the toughening behavior of ferroelectric ceramics with permeable crack face boundary conditions. The influence of electric field on the fracture toughening for conducting cracks is investigated in Chapter 5. The main results of this study are summarized in Chapter 6 and several possible future research directions are also mentioned.
Chapter 2 A Phenomenological Constitutive Model for Ferroelectrics

2.1 Introduction

The need for accurate yet simple constitutive laws for ferroelectric ceramics arises in a number of problems that require the resolution of the inhomogeneous electrical and mechanical fields in ferroelectric materials. Such problems range from the prediction of poling patterns around electrodes in ferroelectric actuator designs to the determination of switching zones near crack tips that govern failure in these materials. Due to the relative complexity of the constitutive response of ferroelectrics, these types of problems will most likely be solved by implementing the finite element method or some other numerical technique. Hence, it is desirable to keep the mathematical description of the constitutive response as simple as possible while maintaining the prominent physical features of the material behavior.

In this chapter, the governing balance equations for ferroelectric materials will be presented first. A general phenomenological formulation for the fully coupled incremental constitutive behavior of ferroelectrics will then be introduced. The foundation of this model is a thermodynamically consistent free energy approach. Micro-electromechanical results for saturation conditions are implemented within the theory to help determine the form of the Helmholtz free energy. Switching surfaces and an associated flow rule for the increments of remanent strain and polarization are proposed to ensure that the energy dissipation rate in the material is always non-negative. In this
chapter various predictions of electromechanical constitutive response such as hysteresis and depolarization behavior will be made and compared to experimental observations.

2.2 Governing equations

The basic equations that govern small deformations and isothermal processes in electromechanical materials will be presented in this section. Standard index notation is utilized with summation implied over repeated indices and the subscript ,j representing differentiation with respect to the xj co-ordinate direction. Consider a volume of ferroelectric material, V, bounded by the surface, S. The mechanical equilibrium equations are given by

\[ \sigma_{ij,j} + b_i = 0 \quad \text{in } V \]  \hspace{1cm} (2.1)

and

\[ \sigma_{ij}n_j = t_i \quad \text{on } S, \]  \hspace{1cm} (2.2)

where \( \sigma_{ij} \) is the Cauchy stress tensor, \( b_i \) is the body force per unit volume, \( n_j \) is a unit vector normal to the surface directed outward from the volume, \( t_i \) is the traction applied to the surface.

The infinitesimal strain \( \varepsilon_{ij} \) is related to displacement \( u_i \) as

\[ \varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}) \]  \hspace{1cm} (2.3)

The electric displacement \( D_i \) satisfies Gauss’ law

\[ D_{i,i} = q \quad \text{in } V \]  \hspace{1cm} (2.4)

and
\[ D n = -\omega \quad \text{on } S \quad (2.5) \]

where \( q \) is the free charge per unit volume in \( V \) and \( \omega \) is the free charge per unit area on the surface.

Under quasi-static conditions the electric field, \( E_i \), can be written as the gradient of the electric potential, \( \phi \), such that

\[ E_i = -\phi_j \quad (2.6) \]

Depending on which independent fields variables are used, there are four forms of the linear piezoelectric constitutive law about a fixed remanent strain and polarization state:

\[
\begin{align*}
\varepsilon_{ij} - \varepsilon^r_{ij} &= S^E_{ijkl} \sigma_{kl} + d_{ijkl} E_k, \quad D_i - P_i^r &= d_{ikl} \sigma_{kl} + \kappa^E_{ij} E_j, \\
\sigma_{ij} &= C_{ijkl}^E (\varepsilon_{kl} - \varepsilon^r_{kl}) - e_{ijkl} E_k, \quad D_i - P_i^r &= e_{ikl} (\varepsilon_{kl} - \varepsilon^r_{kl}) + \kappa^E_{ij} E_j, \\
\sigma_{ij} &= C_{ijkl}^D (\varepsilon_{kl} - \varepsilon^r_{kl}) - h_{ijkl} (D_k - P_k^r), \quad E_i = -h_{ikl} (\varepsilon_{kl} - \varepsilon^r_{kl}) + \beta^E_{ij} (D_j - P_j^r), \\
\varepsilon_{ij} - \varepsilon^r_{ij} &= S^D_{ijkl} \sigma_{kl} + g_{ijkl} (D_k - P_k^r), \quad E_i = -g_{ikl} \sigma_{kl} + \beta^D_{ij} (D_j - P_j^r),
\end{align*}
\]

(2.7) (2.8) (2.9) (2.10)

where \( \varepsilon^r_{ij} \) and \( P^r_i \) are the remanent strain and polarization, \( S^E_{ijkl} \), \( S^D_{ijkl} \), \( C^E_{ijkl} \) and \( C^D_{ijkl} \) are fourth rank tensors of elasticity, \( d_{ijkl} \), \( e_{ijkl} \), \( h_{ijkl} \) and \( g_{ijkl} \) are third rank tensors of piezoelectricity and \( \kappa^E_{ij} \), \( \kappa^D_{ij} \), \( \beta^E_{ij} \) and \( \beta^D_{ij} \) are second rank dielectric tensors. Relationships among the different coefficients can be obtained by algebraically manipulating the different forms of the constitutive law.

Finally, equations (2.1)-(2.6) can be written in the following weak form as

\[ \int_V \sigma_{ij} \delta \varepsilon_{ij} + E_i \delta D_i dV = \int_V b_i \delta u_i + \phi \delta q dV + \int_S t_i \delta u_i + \phi \delta \omega dS \quad (2.11) \]

Equation (2.11) will be the basis for the vector potential finite-element formulation that will be introduced in the following chapter.
2.3 Formulation for the incremental constitutive model

Experimentally, if we increase stresses by \( \Delta\sigma \), or electric fields by \( \Delta E \), the resulting total strain and total electric displacement can be measured without considering how the remanent state in the material changes. Theoretically, suppose that the quantities \( \sigma, E, \varepsilon, D \), \( \varepsilon' \) and \( P' \) are known at time \( t \). If the stresses and electric fields are increased by \( \Delta\sigma \) and \( \Delta E \), in order to determine \( \varepsilon \) and \( D \) at time \( t + \Delta t \), one also needs to predict the changes of remanent state \( \Delta\varepsilon' \) and \( \Delta P' \). In fact, most nonlinear constitutive models, including the one presented in this section for ferroelectric ceramics, are devoted to a formulation that will provide the evolution of the strain, electric displacement, remanent strain and remanent polarization histories given the total stress and electric field histories.

The goal of any phenomenological constitutive theory is to provide a relatively simple framework within which the laws of thermodynamics are satisfied and a wide range of material behaviors can be represented. A summary of the recent developments on micro-electromechanical and phenomenological constitutive modeling of ferroelectrics can be found in review articles by Kamlah (2001) and Landis (2004a). The phenomenological constitutive model presented below is based on the work of Landis and co-workers (2002a, 2003a, 2004b). This constitutive model has been verified against experimental observations and microelectromechanical self-consistent simulations based on the model of Huber et al. (1999).

Similar to Bassiouny et al. (1988) and Cocks and McMeeking (1999), the Helmholtz free energy of the polycrystal contains the reversible or stored part of the free energy and
the remanent state of the material. This assumption is intended to account for the free energy associated with the changes in stored energy due to intergranular constraints that generate internal residual stresses and electric fields during loading. The form of Helmholtz free energy is given as

\[
\psi = \frac{1}{2} C_{ijkl}^D (\epsilon_{ij} - \epsilon_0^r)(\epsilon_{kl} - \epsilon_0^r) - h_{ij} (D_i - P_i^r)(\epsilon_{ij} - \epsilon_0^r) \\
+ \frac{1}{2} \beta_{ij}^e (D_i - P_i^r)(D_j - P_j^r) + \psi^f (\epsilon_{ij}^r, P_i^r)
\]  

(2.12)

where the linear elastic, piezoelectric and dielectric properties of the polycrystal, \( C_{ijkl}^D \), \( h_{ij} \) and \( \beta_{ij}^e \) can each depend on the remanent state of the material. In a polycrystal the linear elastic, dielectric and piezoelectric properties are dependent on the internal variables, i.e., remanent strain and remanent polarization, at each material point. For a material poled by a uniaxial electric field these properties will be homogeneous and transversely isotropic about the poling direction. In comparison to the piezoelectric properties, which must change sign as the direction of the remanent polarization reverses, the elastic and dielectric properties have a much weaker dependence on the remanent state. Hence, for simplicity it will be assumed that elastic compliance at constant electric field, \( S_{ijkl}^E \), and the dielectric permittivity at constant stress, \( \kappa_{ij}^D \), are not affected by changes in the remanent state of the material. The piezoelectric properties are assumed to be linearly dependent on the remanent polarization magnitude and transversely isotropic about the remanent polarization direction. Given these assumptions, the \( S_{ijkl}^E \) and \( \kappa_{ij}^D \) tensors will take on their isotropic forms. However, due to the presence of piezoelectricity and its dependence on the remanent polarization, other elastic and dielectric tensors such as the elastic stiffness at constant electric displacement, \( C_{ijkl}^D \), and
the inverse dielectric permittivity at constant strain, $\beta^e_{ij}$, will have transversely isotropic symmetry about the remanent polarization direction. With these assumptions the theory is greatly simplified and the $S^e_{ijkl}$, $d_{ij}$ and $\kappa^a_{ij}$ tensors are expressed as

$$S^e_{ijkl} = \frac{1}{4\mu} (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \frac{\nu}{2\mu(1+\nu)} \delta_{ij}\delta_{kl} \quad (2.13)$$

$$\kappa^a_{ij} = \kappa \delta_{ij} \quad (2.14)$$

$$d_{ij} = \frac{d_{33}}{4} \frac{P'}{P_0} (3n_j \delta_{ik} + 3n_i \delta_{jk} - 2n_i \delta_{ij}) \quad (2.15)$$

where

$$P' = \sqrt{P'_i P'_i} \quad \text{and} \quad n_i = P'_i / P' \quad (2.16)$$

Here, $\kappa$ is the dielectric permittivity of the material measured at constant applied stress, $\mu$ and $\nu$ are the shear modulus and Poisson’s ratio measured at constant applied electric field, and $d_{33}$ is the piezoelectric coefficient when the material reaches its maximum remanent polarization $P_0$. Note that this form for the piezoelectric coefficients has the additional assumptions that $d_{31} = -d_{33}/2$ and $d_{15} = 3d_{33}/2$. This is a reasonable assumption based on measured values in polycrystals, and this assumption can be relaxed at the added expense of complexity within the theory.

The nonlinear constitutive response of ferroelectric materials is a result of the mechanism of domain switching. In the present theory, domain switching occurs when a specific switching condition is met. This switching criterion can be used to define a surface in stress and electric field space and will be referred to as the switching surface. Within the switching surface the material responds in a linear elastic, piezoelectric and dielectric manner about the current state of remanent strain and remanent polarization.
The second law of thermodynamics implies that the dissipation rate, \( \dot{\Delta} \), for the material is non-negative, i.e.

\[
\dot{\Delta} = \sigma_{ij} \dot{e}_{ij} + E_i \dot{D}_i - \psi \\
= \hat{\sigma}_{ij} \dot{e}_{ij}^r + \hat{E}_i \dot{P}_i^r \geq 0. 
\]  
(2.17)

where the modified stress and electric field are

\[
\hat{\sigma}_{ij} = \sigma_{ij} - \frac{\partial \psi}{\partial \varepsilon_{ij}^r}, \text{ with } \dot{\varepsilon}_{ij} = \frac{1}{3} \sigma_{kk} \delta_{ij} \text{ and } \dot{E}_i = E_i - \frac{\partial \psi}{\partial P_i^r} + \frac{\partial d_{kl}^r}{\partial P_i^r} E_j \sigma_{kl}^r 
\]
(2.18)

where \( \sigma_0 \) is the initial switching strength of the material in uniaxial tension or compression, \( E_0 \) is the coercive field, and \( \beta \) is a positive scalar parameter. The terms \( \hat{\sigma}_{ij} \) and \( \hat{E}_i \) come from the derivatives of the Helmholtz free energy \( \psi \) with respect to \( \varepsilon_{ij}^r \) and \( P_i^r \) and a Legendre transformation is used.

It is possible to satisfy Eq. (2.17) with separate switching surfaces and flow potentials; however, a simpler associated flow rule is utilized here. Specifically, the inequality of Eq. (2.17) and additionally the electromechanical form of the postulate of maximum plastic dissipation, Drucker (1952), will be satisfied for any convex yield surface, \( \Phi \), containing the origin in \( (\hat{\sigma}_{ij}, \hat{E}_i) \) space with an associated normality condition for the flow rule. Note that the switching surface does not necessarily enclose the origin in standard stress and electric field space. The specific form of the switching surface implemented here is that proposed by Landis (2002a):

\[
\Phi = \frac{3}{2} \frac{\hat{\varepsilon}_{ij} \hat{\varepsilon}_{ij}}{\sigma_0^2} + \frac{\hat{E}_i \hat{E}_i}{E_0^2} + \frac{\beta \hat{E}_i \hat{P}_i^r \hat{P}_i^r}{E_0 P_0^r \sigma_0^2} - 1 = 0 
\]
(2.19)
The switching surface defined in Eq. (2.19) must be satisfied during switching and is convex if $\beta < 3$. Normality condition requires that remanent increments of remanent strain and polarization are normal to the switching surface such that

$$\Delta e'_{\theta} = \lambda \frac{\partial \Phi}{\partial \sigma_{\theta}} \quad \text{and} \quad \Delta P' = \lambda \frac{\partial \Phi}{\partial E},$$

(2.20)

where $\lambda$ is the switching multiplier.

Finally, the ten equations in (2.19) and (2.20) can be solved for the unknowns $\lambda$, $\Delta e'_{\theta}$, and $\Delta P'$ if the form of $\psi'$ is specified to complete the constitutive theory. For the current theory, $\psi'$ is additively decomposed into a mechanical part $\psi^m$ that enforces the strain saturation conditions, and an electrical part $\psi^e$ that enforces the polarization saturation conditions, i.e.,

$$\psi' = \psi^m + \psi^e$$

(2.20)

$$\psi^m = \frac{1}{2} H_0^m \left[ \frac{J^r_{\theta}}{E_c} \exp \left( \frac{m}{1 - \bar{\varepsilon}/E_c} \right) \right]^2$$

(2.21)

where $H_0^m$ is a characteristic level of back stress that primarily affects the initial slope of uniaxial stress versus remanent strain curve, and $m$ is another hardening parameter that controls how abruptly the strain saturation conditions can be approached. The multi-axial remanent strain saturation conditions are enforced by causing $\psi^m$ to approach infinity as the strain-like variable $\bar{\varepsilon}$ approaches the saturation level of remanent strain in uniaxial compression $E_c$. The effective saturation remanent strain quantity $\bar{\varepsilon}$ is defined as

$$\bar{\varepsilon} = J^s f \left( J^r_{\theta}/J^s \right)$$

(2.22)

where
\[ f \left( \frac{J^e_3}{J^e_2} \right) = -0.0965 \left( \frac{J^e_3}{J^e_2} \right)^3 + 0.01 \left( \frac{J^e_3}{J^e_2} \right)^6 + 0.8935 \text{ for } \left( \frac{J^e_3}{J^e_2} \right) < 0, \quad (2.23) \]

and

\[ f \left( \frac{J^e_3}{J^e_2} \right) = -0.1075 \left( \frac{J^e_3}{J^e_2} \right)^3 - 0.027 \left( \frac{J^e_3}{J^e_2} \right)^6 - 0.028 \left( \frac{J^e_3}{J^e_2} \right)^{21} + 0.8935 \text{ for } \left( \frac{J^e_3}{J^e_2} \right) \geq 0. \quad (2.24) \]

Here, \( f \) is a functional fit to the numerical results obtained from the micromechanical computations described in Landis (2003a). The following remanent strain invariants are used to describe the multi-axial remanent strain state

\[ J^r_2 = \left( \frac{2}{3} e''_{ij} e'_{ij} \right)^{\frac{1}{2}} \text{ and } J^r_3 = \left( \frac{4}{3} e''_{ij} e'_{ij} e'_{jk} \right)^{\frac{1}{3}}. \quad (2.25) \]

where \( e'_{ij} \) is the remanent strain deviator, \( e''_{ij} = e'_{ij} - \delta_{ij} e'_{kk} / 3 \).

Next, the electrical part of \( \psi' \) has the form of

\[ \psi' = H_0^E P_0 \left[ \ln \left( \frac{1}{1 - P'/P_{sat}} \right) - \frac{P'}{P_{sat}} \right], \quad (2.26) \]

where

\[ P_{sat} = \frac{3P_0}{4(\epsilon_c + \epsilon_e)}(e''_{ij}n_i n_j + \epsilon_e) + \frac{P_0}{4} \quad (2.27) \]

Here \( \epsilon_r \) is the remanent saturation strain in uniaxial tension and according to Eqs. (2.21)-(2.24) is equal to 1.368\( \epsilon_r \). The hardening parameter \( H_0^E \) is a characteristic level of back electric field that primarily affects the slope of the uniaxial electric field versus remanent polarization response. The maximum attainable remanent polarization \( P_0 \) has been defined previously. Note that the level where the remanent polarization saturates \( P_{sat} \) is a function of the remanent strain and the maximum remanent polarization level of \( P_0 \) can only be attained if \( e''_{ij}n_i n_j = \epsilon_e \). If \( e''_{ij}n_i n_j = -\epsilon_e \), then the maximum level for \( P' \) is only
$P_0/4$. This result and the linear approximation to the functional form for $P_{sat}$ given in Eq. (2.27) are taken directly from the micro-electromechanical computations described in Landis et al. (2004b).

### 2.4 Results

In this section, the above fully coupled theory is solved by using the Newton-Raphson method described in Appendix A. The simulation results are explained and compared with the experimental measurements of Lynch (1996) and Huber and Fleck (2001). The material properties and constitutive parameters to be used are characteristic of a soft PLZT material as measured by Lynch, and are specifically given as: $\sigma_0 = 27.5$MPa, $E_0 = 0.35$MV/m, $P_0 = 0.26$C/m$^2$, $\varepsilon_\infty = 0.12\%$, $\beta = 2.95$, $\kappa = 6 \times 10^{-8} C/(m \cdot V)$, $E = 70$GPa, $\nu = 0.4$, $d_{33} = 3 \times 10^{-10}$ m/V, $m = 0.01$, $H_0^\sigma = 0.5\sigma_0$, $H_0^E = 0.05E_0$.

#### 2.4.1 Pure electrical loading

The first test of the theory is the response of the ferroelectric material under purely electrical loading. Figure 2.1 shows the theoretical prediction of the typical hysteresis response of ferroelectrics under uniaxial electrical loading. At zero electric field, the polarization vectors in the domains are randomly distributed as shown in the schematic domain structure for the state A, and the macroscopic average of polarization is zero. When the electric field is gradually increased but does not reach the coercive field, the negative and positive charges of the unit cell are only slightly displaced from their equilibrium position. For this reason, the configuration of the domains is not altered and the macroscopic dielectric response of the material is linear and reversible. If the electric
field is reduced back to zero, the material will return to the initial state. The domain switching processes are triggered as soon as the electric field reaches the coercive field $E_0$. A slight increase of the electric field will cause the polarization to climb dramatically, as the polarization vectors in the domains are switched toward the direction of the applied increasing electric field. However, due to the constraints of the material structure, the polarization cannot increase without limit. Instead, a saturation state will be reached in the material when all domains have switched towards the electric field. A further increase of the electric field will only introduce linear changes of the polarization. State B in Figure 2.1 shows the saturation state when the switching of most of the domains is complete.

Figure 2.1: Prediction of the electric displacement versus electric field hysteresis loop under uniaxial electric field loading at zero stress. The domain structures at states A-G are for the purpose of explanation.
If the electric field is unloaded from any value higher than the coercive field, many domains remain in their switched state, and the poled domain structure is conserved as shown in Figure 1.5. In this irreversible process, the material shows a macroscopic remanent polarization even when the electric field is totally taken away (see the state C in Figure 2.1 in this case). If the electric field is then applied in the reverse direction, the polarization is gradually switched back and will decrease to zero when the coercive field has arrived at state D. Further reversal of the electric field switches the polarization to align in the direction of the applied electric field, and finally the material reaches the saturation state (E in Figure 2.1) again. The response of the material along the loading path E-F-G-B in Figure 2.1 is then similar to B-C-D-E path.

The hysteresis loop in Figure 2.1 has several characteristics that are worth restating here: The initial response of the material is a linear and reversible process. Irreversible switching processes happen when the load reaches the coercive field. The polarization approaches saturation with the increase of the electric field. The hysteresis occurs due to the irreversibility of the domain switching process.

Corresponding to Figure 2.1 is the change of the strain of the specimen during the electric loading cycle as shown in Figure 2.2. The corresponding letters in both figures indicate the same material state. Notice that no macroscopic strain is found when the electric field is less than coercive value. The reason for this behavior is due to the arbitrarily dispersed polarization vectors which cause the microscopic piezoelectric strain contributions of the individual domains to average to zero. State A in Figure 2.2 where the stain is zero and the material is isotropic is called the thermally depoled state. The
switching process starts when the applied electric field exceeds $E_o$. Similar to the rapid increase of polarization, the sample is elongated dramatically with the increase of electric field above $E_o$. As shown in the governing equations, the growth of the total strain is due to its two decomposed parts: the accumulating remanent strain and the macroscopic linear piezoelectric effect due to the remanent polarization. From Figure 1.3 we can see that the polarization vector is parallel to the c-axes or longest axes of the unit cell. More unit cells switching towards the direction of the electric field implies more of c-axes aligned in the direction of the electric field. Finally the switching process is completed and the material reaches the saturation state when state B in Figure 2.2 is approached.

![Figure 2.2: Prediction of the strain versus electric field butterfly loop corresponding to the hysteresis loop under uniaxial electric loading at zero stress. States A-G here are the same as those in Figure 2.1.](image)

27
After saturation, the remanent strain remains unchanged and the contribution to the strain arises only from the linear piezoelectric effect. Linear piezoelectric unloading also occurs from state B to C in Figure 2.2. The macroscopic remanent strain is the average effect of the alignment of the c-axes of the domains. If the electric field is reversed, the polarization distribution becomes more and more random and so does the c-axes distribution thus reducing the macroscopic strain. The strain arrives the minimum value at state D when the macroscopic polarization reduces to zero. Here the minimum strain at state D is positive but in other materials it may actually be negative.

When the electric field is further reversed beyond the negative coercive field, the switching of the polarization towards the direction of the applied electric field begins again and saturation occurs near state E. The loading path of E-F-G-B has similar switching behavior as B-C-D-E, though the direction is opposite. As seen from Figure 2.2, both high negative and positive electric field causes positive strains, resulting in symmetry of the strain with respect to zero electric field. In fact, from Figure 1.3, we can see that positive and negative electric fields can cause the polarization vector to point to opposite directions, but the c-axes of the unit cell will always remain in the line of the direction of the electric field. As a result, the macroscopic average of the unit cells in the material shows an elongation effect for both negative and positive electric field loadings.

2.4.2 Purely mechanical loading

In this section the simulation results for a ferroelectric material subjected to purely mechanical loading under zero electric field is discussed. Starting with a small tensile loading from a thermally poled state A in Figure 2.3, domain switching is not induced
and the material shows linear elastic behavior. After passing the switching strength $\sigma_0$, domain switching processes are triggered. With the increase of stress, domains are switched such that the c-axes of the unit cells are aligned in the direction of the tensile loading. As seen from the simulation results, the strain increases but no polarization is found. This phenomenon can be explained from the micro-structural response of the material in Figure 1.6. For the polarization parallel to the loading, the tensile stress can only stretch the unit cell along the line of loading without changing the polarization direction. If the polarization is perpendicular to the tensile stress, the energetics for switching the polarization into the negative and positive directions are equal. As a result, if the material does not possess a macroscopic polarization initially, the polarization switching induced by the mechanical loading will have a macroscopic average that shows no net polarization. Similar to the dielectric behavior, the material reaches a fully switched state or saturation state at a high level of applied stress (around $2\sigma_0$ in this study). When the stress is further increased, the material responds linearly before reaching state B. Unloading from B to C does not cause significant reverse switching, which leaves a large remanent strain state when the stress is fully released.

If the stress is then reversed (compressive stress) from state C, domain switching happens immediately. However, since the opportunities of switching the polarization into the four directions perpendicular to the loading direction are the same for a unit cell, similar to the case in tension, no macroscopic average polarization is introduced in the polycrystalline ferroelectrics by the compressive stress. In order to recover the material to its original undeformed state, a magnitude of coercive value $-\sigma_0$ needs to be applied. Reversal of the original switching happens during the loading from C to D. Further
increase of the compressive stress will induce negative strains and the reverse process will happen in a similar way along the path E-F-G.

![Graph](image)

Figure 2.3: Simulation of ferroelastic behavior: uniaxial stress versus longitudinal strain hysteresis loop at zero electric field.

Notice in Figure 2.3 that the absolute values of strain at states C and F are not equal. This asymmetry under tensile and compressive stress is one of the important features of ferroelastic behavior of the ferroelectric ceramics. It is not difficult to see that the remanent strains under tensile and compressive loadings are not the same. From simple considerations (Sheng, 2003) of a single BaTiO$_3$ crystal which contains a set of parallel unit cells in Figure 1.1, it is shown that the ratio between maximum remanent strain in tension and maximum switching strain in compression can be up to 2. However in this
study, the calculation is conducted on a polycrystal with a random distribution of the
orientation of its grains. According to the microscopic studies in Landis (2003a) and
Landis et al. (2004b), the ratio between maximum tensile and compressive remanent
strains in the polycrystal ferroelectric ceramics is around 1.367:1, much smaller than the
2:1 from the aforementioned single crystal consideration.

2.4.3 Combined electromechanical loadings

Since the ferroelectric devices often operate in electromechanical environments,
experiments have been carried out to study the material response to complex loadings
(Lynch, 1996). Here we will show the model results for mechanical depolarization and
electric field cycling with prescribed compressive stress on ferroelectric materials.

![Graph showing stress versus electric displacement](image)

Figure 2.4: Stress versus electric displacement during the simulation of mechanical
depolarization of poled specimen.
Mechanical depolarization happens when a prepoled material is subjected to a compressive stress parallel to the direction of the polarization. To simulate this situation, the specimen is first electrically loaded to an amplitude of $E = 3E_0$ under zero stress state. The electric field is then removed and a compressive stress parallel to the original electric field is then applied. During the electromechanical loadings, the changes of polarization and strain are recorded simultaneously. Since the material response to electric poling under the condition of stress free state is discussed in section 2.4.1, only the response during the mechanical depolarization is discussed below.

Figure 2.4 shows the relationship between the polarization and the stress during the mechanical depolarization. With a macroscopic polarization in the material, unlike the case of mechanical loading with no electrical poling, even small compressive loading can cause domain switching. With the increase of the stress, more and more polarization vectors are switched towards the plane that is perpendicular to the loading direction. As a result, the initial polarization induced by the electrical poling decreases gradually. However, macroscopically the material does not show polarization in the directions perpendicular to the compressive stress since the loading induces an axisymmetric state. When the polarization is decreased to a certain level, further change of the polarization becomes very slow even with high stress. In fact, the polarization cannot be reduced to zero simply by the compressive stress due to the orientations of the grains and material structure. The unloading curve is nearly a straight line and the polarization is only recovered a little bit.

Corresponding to the mechanical depolarization behavior, the changes of longitudinal
strain are recorded in Figure 2.5. Comparing Figure 2.5 and Figure 2.3, we can find that the strain responses in these two compressive loadings are very similar. In both cases, the initial elongated material contracts, returns to the undeformed state, becomes compressed and then reaches saturation as the compressive load is increased; the unloading curves both follow a linear elastic response and display a small amount of non-linearity at the end. The inherent difference between the purely ferroelastic behavior and mechanical

![Graph showing stress versus strain](image)

Figure 2.5: The counterpart of Figure 2.4: stress versus strain during the simulation of mechanical depolarization of poled specimen.

depolarization is the initial state. In purely mechanical loading, the initial remanent strain is induced by the mechanical load and no macroscopic polarization is created. On the other hand, the electrical loading before mechanical depolarization leaves both a remanent strain and a macroscopic polarization. The macroscopic polarization is reduced during the mechanical loading but is present even after the unloading of the compressive
stress. In fact, the complete removal of the polarization can only be achieved by applying an electric field in its opposite direction.

In addition to hysteresis, butterfly, stress-strain and depolarization loops, Lynch (1996) also measured hysteresis and butterfly loops for material under a fixed compression. Figure 2.6 shows the model predictions for hysteresis and butterfly loops under the application of a constant applied compressive stress. The loading procedure is as follows. Initially, the material is poled by a strong electric field \(3E_0\) and the field is removed. Next, a compressive stress is applied and held constant at a given level. The constant superimposed stress levels range from 0 to \(-3\sigma_0\) in these simulations. Finally, the electric field is cycled between \(-3E_0\) and \(3E_0\) beginning with the electric field applied in the negative direction and the resulting hysteresis and butterfly loops are monitored. During the total loading cycle, the electric field and compressive stress are along the same axis. Notice the points labeled A, B, and C in Figure 2.4 and Figure 2.5. These points correspond to the hysteresis and butterfly loops labeled A, B and C in Figure 2.6.

The most noticeable feature in Figure 2.6 is that both the hysteresis and butterfly loops become thinner with the increase of the constant applied compressive stress. After the material is poled to \(3E_0\) and then unloaded to zero electric field under stress free state, the material is left with a remanent polarization in the direction of the poling field. The application of compressive stress induces a 90 degree switching process and decreases the remanent strain and remanent polarization caused by the previous electrical poling. However, the electric field applied after the constant compressive stress wants to align the domains in the direction of the electric field while the compressive stress tends
Figure 2.6: Predictions of polarization versus electric field and strain versus electric field hysteresis loops under different constant preloaded compressive stress. The initial states in (A), (B), and (C) correspond to the state of A, B, and C in Figure 2.4.
to switch the domains into the direction perpendicular to the electric field. Thus the
electric field and compressive stress compete with each other to switch the domains into
their favorite direction. As a result, unlike the sharp jump around the coercive field in
stress free situation, the changes of strain and polarization are a slower process. Also with
the increase of compressive stress, more and more domains are switched toward the
direction perpendicular to the stress and the stress makes the possible maximum remanent
strain and maximum remanent polarization smaller. Fewer and fewer domains take part
in the thereafter electrical switching and the consequential hysteresis and butterfly loops
turn out to be flatter.

2.4.4 Multi-axial polarization rotation tests

The final set of constitutive law simulations pertain to the multi-axial polarization
rotation experiments of Huber and Fleck (2001) which are simulated within the theory
and are plotted in Figures 2.7. The experiment is performed as follows. Initially, an
unpoled specimen is poled by a strong electric field. Then, a new specimen is cut and
coated with electrode material such that the initial polarization direction is at an angle of
$\theta$ to the new electric field direction which is perpendicular to the electrodes. An electric
field of magnitude $3E_0$ is applied to the specimen and the electric displacement change
$\Delta D$ and axial strain change $\Delta \varepsilon$ in the direction of the applied electric field are measured.
In the original experiments by Huber and Fleck only the electric displacement changes
were measured. The qualitative agreement between the simulations displayed on the left
in Figure 2.7 and measurements of Huber and Fleck is exceptional. The corresponding
strain changes plotted in Figure 2.7 are included as possible predictions for future experimental observations of strain for this type of experiment.

Figure 2.7: Polarization rotation tests obtained from the constitutive law. Left: Change of the electric displacement vs. electric field. Right: Change of strain vs. electric field.

When the initial polarization direction is at an angle of $\theta$ to the new electric field direction, the polarization can be decomposed into two parts: one parallel and the other perpendicular to the electric field. The new electric field tends to induce the 90 degree switching to the perpendicular polarization and the 180 degree switching to the parallel polarization if the parallel polarization is in the opposite direction of the electric field. However, if the polarization is in the same direction of the electric field, the electric field cannot affect those domains. From the above analysis, we know that the new electric field can cause little changes of polarization at 0 degree, much bigger changes of polarization are made at $\theta = 180^0$ and in all other cases the changes of polarization are between those at $\theta = 0^0$ and $\theta = 180^0$. The strain change is a little more complex but we would expect that the electric field would cause the highest change at $\theta = 90^0$ since the switchable
domains are the most available in this case. Also in Figure 2.7 the changes of strain first go down and then climb up again for the cases of $90^0 < \theta \leq 180^0$. The reason is the electric field needs to back switching the domains first and then align the domains in the new direction.

2.5 Summary

In this chapter a micro-electromechanically based phenomenological model for the constitutive behavior of polycrystalline ferroelectrics was proposed. The constitutive law presented is an internal variable plasticity theory. The internal variables that define the state of the material are the remanent polarization and the remanent strain. It is assumed that the internal state of the material is uniquely defined by these variables. Associated with the internal variables is a Helmholtz free energy potential from which hardening moduli are readily obtained from the invariants of the internal state variables. The saturation information obtained from micro-electromechanical simulations has been used to determine features of the remanent potential within the phenomenological framework in order to ensure that the model can not produce unattainable combinations of remanent strain and remanent polarization. The predictions of the phenomenological model are found to be in excellent qualitative accord with experimental observations of polycrystalline material behavior. In the next chapters, this constitutive model will be implemented into a new finite element method to study the steady state fracture behavior of ferroelectric materials under different loading conditions.
Chapter 3 The Fracture Toughness of Ferroelectric Ceramics with an Electric Field Applied Parallel to the Crack Front

3.1 Introduction

The work in this chapter is motivated by recent experimental and theoretical investigations on the effects of electric fields on the fracture toughness of ferroelectrics. The investigations presented here have focused on the specific case where the electric polarization and electric field in the sample are in the out-of-plane direction parallel to the crack front as illustrated in Figure 3.1. Meschke et al. (1997) and Kolleck et al. (2000) have observed experimentally that the steady-state fracture toughness of ferroelectrics increases with the increase of applied electric field to an initially unpolarized specimen. Additionally, Lucato et al. (2002) and Hackemann and Pfeiffer (2003) have observed that out-of-plane poling has practically no effect on the fracture toughness of ferroelectrics when the out-of-plane electric field is zero.

Recent theoretical explanations of the experimental phenomena have focused on the role of domain switching in the toughening process. Kolleck et al. (2000) and Yang et al. (2001) treat domain switching with the concepts developed for transformation toughening in partially stabilized zirconia (McMeeking and Evans, 1982; Budiansky et al., 1983) to analyze the fracture toughening behavior. Kreher (2002) proposed a fracture model based on the balance of energy supplied by the driving forces and the total energy either dissipated by domain switching, stored in the crack wake region or consumed by the
formation of new fracture surface. In these proposed theories either the details of the mechanical and electrical coupling behavior of the material are neglected or the calculation of the complicated crack tip electromechanical fields is avoided. The model to be presented in this chapter amends these simplifications by applying the

![Diagram of electromechanical configuration](image)

Figure 3.1: A schematic of the out-of-plane electrical and in-plane mechanical configuration to be modeled in this work. For any given sample, the electric field and remanent polarization are aligned in \( x_2 \) direction. The in-plane mechanical loading is simply indicative of the Mode I symmetry to be modeled in this work and should not be interpreted literally.

phenomenological constitutive law developed in Chapter 2 for ferroelectric switching within the finite element method to determine the details of the crack tip fields and the toughening due to domain switching during steady crack growth.

The remainder of this chapter follows from the published paper by Wang and Landis (2004) and is organized as follows. In Section 3.2 the nonlinear phenomenological
constitutive model developed in Chapter 2 is tailored for the specific case with electric effect parallel to the crack front only. The fracture model and the finite element formulation which implements the constitutive model for steady crack growth are also described in this section. The results will be presented and analyzed in Section 3.3. A discussion of the results and comparisons to experimental observations is included in Section 3.4.

3.2 The fracture model and finite element formulation

In this work the primary mechanical loading is applied in the $x_1$-$x_2$ plane and electric field is applied only in the $x_3$ direction as illustrated in Figure 3.1. Due to the constraint of plane-strain, axial stresses in the $x_3$ direction are allowed to develop as well. Also, due to symmetry of the loading, remanent polarization can develop only in the $x_3$ direction. From the discussion of the material properties in Chapter 2, we know that materials poled in the $x_3$ direction will have elastic, dielectric and piezoelectric properties that are transversely isotropic with the $x_1$-$x_2$ plane being the plane of isotropy. The elastic and dielectric properties are not affected by changes in the remanent polarization of the material. Also the piezoelectric properties are proportional to the remanent polarization. Under these conditions, the constitutive relationships (2.7) can be reduced to

$$
\varepsilon_{ij} = \frac{1 + \nu}{E} \sigma_{ij} - \frac{\nu}{E} \sigma_{kk} \delta_{ij} + (\varepsilon_{ij}^r - \varepsilon_{ij}^0) + d_{3ij} E_3 \quad (3.1)
$$

$$
D_3 = d_{3ij} \sigma_{ij} + \kappa E_3 + (P_3^r - D_3^0) \quad (3.2)
$$

Here, $\varepsilon_{ij}$ are the Cartesian components of the infinitesimal strain tensor as referenced from the initial strain state $\varepsilon_{ij}^0$. The components of the remanent strain as referenced from
a thermally depolarized state are given as \( \epsilon_{ij}^{\prime} \). Furthermore, \( d_{ij}^{\prime} \) are the components of the piezoelectric tensor, \( E_3 \) is the electric field in the \( x_3 \) direction, \( D_3 \) is the electric displacement in \( x_3 \) direction as referenced from the initial electrical displacement \( D_3^0 \), and \( P_3^r \) is the total remanent polarization in \( x_3 \) direction as referenced from the thermally depolarized state.

Eqs. (2.26) and (2.27) can be reduced to

\[
\Psi^E = H_0^{E} \frac{P}{P_0} \left[ \ln \left( \frac{1}{1-P_r^{\prime} \sqrt{P_{sat}}} \right) - \frac{P_{3 \prime}}{P_{sat}} \right]
\]

(3.3)

and

\[
P_{sat} = \frac{3P_0 (\epsilon_{33} + \epsilon_{ij}) + P_0}{4(\epsilon_{ij} + \epsilon_{ij})}
\]

(3.4)

Note that the simplifications described are valid only when only the \( P_3^r \) component of the remanent polarization is involved.

### 3.2.1 The loading process

Prior to crack growth the material is loaded by a uniaxial electric field \( E_3^A \) and there is no mechanical loading involved. After this initial electrical loading operation the initial strain and electric displacement are given as

\[
\epsilon_{ij}^0 = \epsilon_{ij}^{r0} + d_{ij}^0 E_3^A
\]

(3.5)

\[
D_3^0 = P_3^{r0} + \kappa E_3^A
\]

(3.6)

Here, \( \epsilon_{ij}^{r0} \) and \( P_3^{r0} \) are the Cartesian components of the initial remanent strain and initial remanent polarization, and \( d_{ij}^0 \) are the components of the piezoelectric tensor when \( P_3^r = P_3^{r0} \). Note that in all cases discussed here, the reference states for \( \epsilon_{ij}^{\prime} \), \( \epsilon_{ij}^{r0} \), \( P_3^r \), and
$P_{3}^{0}$ correspond to the state of the material as cooled from above the Curie temperature, i.e., a thermally depolarized sample. Within the implementation of the constitutive law, the knowledge of this pre-poled state $\varepsilon_{ij}^{r,0}$ and $P_{3}^{r,0}$ must always be retained and incorporated into the total remanent strain and the total remanent polarization in order to refer to the thermally depolarized state.

For the geometry and loading to be modeled here in Figure 3.1, two types of initial electrical states will be considered: initially unpoled and initially poled states. The initially unpoled samples begin in the thermally depolarized state of the material. The initially poled samples are poled by applying a uniaxial electric field in the $x_{3}$ direction to a level of $E_{3}^{p}$ and then removing the applied field. Note that the poling field $E_{3}^{p}$ must be greater than the coercive field $E_{o}$ in order to induce remanent polarization. This entire electrical loading procedure is performed in the absence of mechanical stress. The poling process induces both residual remanent polarization and strain in the material as referenced from the thermally depolarized state. Figure 3.2 illustrates the (a) electric displacement versus electric field, (b) strain versus electric field response during such electrical loading, (c) depolarization due to mechanical stress, and (d) the stress versus strain response during depolarization. Due to the irreversibility of the domain switching process, there is a continuous range of partial poling states that the material can attain in the range from the initially unpoled state to a fully poled state. Note that the straight lines within the loops in Figures 3.2a and 3.2b represent linear unloading during the removal of the applied electric field, and those in 3.2c and 3.2d depict the initial behavior during
Figure 3.2: The uniaxial electromechanical behavior of the model material with three levels of the poling field $E_3^p$ leading to different partially poled states. (a) The electric field versus electrical displacement hysteresis loops. (b) The electric field versus strain butterfly loop. (c) The stress versus electrical displacement depolarization loop. (d) The stress versus strain loop during depolarization. Notice that the intermediate lines in (a) and (b) represent the response during the unloading of electric field, and those in (c) and (d) represent the depolarization behavior from a partially poled state.

depolarization by compressive stress from different partially poled states. After the initial poling step or lack of it, the electromechanical loading history for the specimen is as
follows. An electric field is applied in the \( x_3 \) direction in the absence of mechanical stress. If the applied electric field is of sufficient magnitude, then poling of initially unpoled samples or a reversal of poling in initially poled samples may result. In any case, this step in the electrical loading procedure induces new states of strain and electric displacement, which have been previously called the initial strain \( \varepsilon_{ij}^0 \) and initial electric displacement \( D_3^0 \). The initial strain and electric displacement consist of both linear reversible parts and remanent parts as given by Equations (3.5) and (3.6). The final step in the loading process is to apply the in-plane mechanical loads while keeping the out-of-plane electric field fixed at the level attained in the previous step. Under plane-strain conditions the out-of-plane axial strain \( \varepsilon_{33} \) is assumed to remain unchanged from its state after the electrical loading step, i.e. \( \varepsilon_{33} = \varepsilon_{33}^0 \). Steady crack growth then occurs while the in-plane mechanical loads are applied.

### 3.2.2 Small scale switching

During crack growth, small scale switching will be assumed such that the representative height of the nonlinear switching zone near the crack tip is much smaller than any other characteristic specimen dimension such as crack length, specimen width or ligament width. Furthermore, under plane-strain conditions it is assumed that the specimen thickness is much greater than the switching zone size as well. The assumption of small scale switching will not be valid when the applied out-of-plane electric field is of sufficient magnitude to cause switching itself. However, it is assumed here that the in-plane applied mechanical loads can still be characterized by a Mode 1 \( K \)-field. Under these conditions a characteristic switching zone half-height, \( R_s \), can be identified as
\[ R_s = \frac{1}{3\pi} \left( \frac{K_1}{\sigma_0} \right)^2 = \frac{1}{3\pi} \frac{G}{E'} \]  

(3.7)

Here \( K_1 \) is the mode I stress intensity factor such that within the small scale switching approximation, the stress around the crack are given as

\[
\begin{bmatrix}
\sigma_{11} \\
\sigma_{22} \\
\sigma_{12}
\end{bmatrix} \to \frac{K_1}{\sqrt{2\pi r}} \begin{bmatrix}
\theta \\
\frac{\theta^2 - \sin 2\theta}{2} \\
\frac{\theta^2 + \sin 2\theta}{2} \\
\frac{\sin \theta}{2}
\end{bmatrix}
\begin{bmatrix}
1 - \sin 2\theta \\
\frac{\theta^2 - 2\sin 3\theta}{2} \\
\theta^2 + \sin 2\theta \\
\frac{\sin \theta}{2}
\end{bmatrix}
\]  

(3.8)

as \( r \to \infty \)

where \( r \) and \( \theta \) represent a polar coordinate system centered on the crack tip, with \( \theta \) measuring the angle between the radial direction and the \( x_1 \)-axis. The expression of (3.8) is applied as far-field boundary conditions and the crack surface is assumed to be traction free. Under small scale switching conditions the prevailing mechanical conditions, which govern the nonlinear behavior near the crack tip due to the geometry and far field loading of a sample, are completely characterized by \( K_1 \). Furthermore, the applied energy release rate, \( G \), is related to \( K_1 \) as

\[ G = \frac{K_1^2}{E'} \]  

(3.9)

where \( E' = E \) for plane-stress and \( E' = \frac{E}{1 - v^2} \) for plane-strain. Note that \( R_s \) is a very good estimate of the switching zone size when there is no out-of-plane applied electric field. However, due to the electromechanical coupling in ferroelectrics, the out-of-plane electric field will affect the in-plane stresses at which switching will occur and consequently the size of the switching zone. This effect will be discussed in further detail in the Results section of this chapter.
It is also important to note that when the applied electric field is near or equal to the coercive field, the assumptions of small scale switching are not rigorously valid. When the applied electric field is equal to \( E_0 \), Equation (2.17) indicates that the switching surface associated with mechanical loading shrinks to a point for unpoled materials. Hence, under such conditions any non-hydrostatic applied stress will cause small increments of switching. However, switching will be relatively diffuse far from the crack tip and it will be assumed that the tractions associated with the \( K \)-field given by Equation (3.8) and the applied energy release rate given by Equation (3.9) will remain approximately valid.

The analysis presented here focuses only on the toughening due to domain switching during the steady crack growth conditions described above. Under steady growth conditions, all increments of field quantities can be related to derivatives with respect to the \( x_i \) coordinate direction by

\[
\dot{\chi} = -\dot{a} \frac{\partial \chi}{\partial x_i} \tag{3.10}
\]

Here, \( \chi \) is any scalar field quantity such as a Cartesian component of remanent strain or remanent polarization. Since the constitutive model used in this study is rate independent, \( \dot{a} \) represents the increment of crack advance in the \( x_i \) direction. If rate dependent material behavior were considered, then \( \dot{a} \) would represent the crack growth rate (Landis et al., 2000). In either case, if inertial effects are neglected, the determination of the energy flux to the crack tip given by Equation (3.11) and the finite element formulation to solve for the electromechanical fields derived from Equations (3.14) are the same. Only the integration of the constitutive law would change with the additional feature of rate dependence.
Within this model, crack propagation will be assumed to occur when the crack tip energy release rate $G_{tip}$ reaches a critical value. To compute the relationship between the maximum or steady state far field applied energy release rate $G_{ss}$ and $G_{tip}$, a steady state finite element formulation will be implemented. In the far field, traction corresponding to Equation (3.8) is applied to the model and Equation (3.9) can be used to compute $G_{ss}$. Then, under steady-state conditions, $G_{tip}$ can be calculated using a formula similar to Hutchinson's $I$-integral (Hutchinson, 1974) as

$$G_{tip} = I = \int_S \left( W n_i - \sigma_{ij} n_j u_{i,i} + D_i n_i E_i \right) dS$$  \hspace{1cm} (3.11)

where $S$ is a surface enclosing the crack tip, $n_i$ are the components of the unit normal directed outward from the surface, $u_i$ are the components of the displacement vector, $D_i$ are the components of the electric displacement vector, $E_i$ is the electric field in $x_1$ direction, and $W$ is the history dependent electric enthalpy density at a material point defined by

$$W = \int_0^{\varepsilon_{s,E_i}} \sigma_{ij} d\varepsilon_{ij} - D_i dE_i$$  \hspace{1cm} (3.12)

In general Equation (3.11) is a surface integral instead of a contour integral because electrical energy can enter the system from the electrodes attached to the surface of the specimen. However, because $E_1 = E_2 = 0$ and $E_3$ is constant throughout the calculation, Equation (3.11) can be simplified to a contour integral as

$$G_{tip} = \int_{\Gamma} (W n_i - \sigma_{ij} n_j u_{i,i}) d\Gamma$$  \hspace{1cm} (3.13)

For a traction-free crack, the contour $\Gamma$ begins on the lower crack face, encircles the crack tip in the counterclockwise sense, and ends on the upper crack face. The calculation of $G_{tip}$ is carried out after the finite element solution is obtained.
3.2.3 Finite element formulation

The finite element formulation required to solve the steady crack growth boundary value problem is based on the variational statement

\[ \int_V \delta e_{ij} C_{ijkl} e^{rs}_{kl} dV = \int_S \delta u_i T_i dS + \int_V \delta e_{ij} C_{ijkl} (e^{rs}_{kl} - e^{rs}_{kl} + d^{rs}_{kl} E_3) dV \]

(3.14)

where $S$ is the boundary of the volume $V$. $C_{ijkl}$ are the Cartesian components of the isotropic elastic stiffness tensor that can be written in terms of $E$ and $v$, and the tractions acting on the boundary $S$ are given as $T_i = \sigma_{ij} n_j$. These tractions are determined from the stress field of Equation (3.8). Note that no electrical terms are needed in this finite element formulation because the electrical field equations, $\nabla \cdot D = 0$ and $\nabla \times E = 0$, are automatically satisfied by the fields $E_1 = E_2 = 0$ and $E_3 = \text{constant}$ with $D_1 = D_2 = 0$ and $D_3 = d_{3j} \sigma_{ij} + \kappa E_3 + P'_3 = D_3(x, y)$. However, the electrical behavior, in particular the changes in $P'_3$ and $d_{3j}$, does have a significant effect on the in-plane mechanical fields through the constitutive behavior of the material. Returning to the finite element formulation, after the application of the appropriate finite element interpolations and the cancellation of the appropriate variational terms, the left-hand side of Equation (3.14) represents the linear elastic stiffness matrix dotted with the vector of unknown nodal displacements in the $n+1^{th}$ iteration. Note that this stiffness matrix does not depend on any non-linear deformations or non-linear polarization, only on the finite element mesh geometry and elastic properties of the material. Hence the stiffness matrix remains constant for all iterations.

The first term on the right-hand side of Equation (3.14) represents the vector of known applied nodal forces arising from the tractions described by Equation (3.8), which
correspond to a specified level of the far field applied energy release rate. Note that these applied tractions do not change from iteration to iteration. Finally, the second term on the right-hand side can be viewed as the body force due to the distributions of piezoelectric strain and remanent strain in the material from the \( n \)\(^{th} \) iteration. As alluded to in this discussion, the finite element Equation (3.14) is solved with an iterative technique. To start, uniform remanent strain and polarization distributions are assumed and integrated on the right hand side of Equation (3.14) and added to the applied traction boundary conditions. Next, the system of finite element equations is solved to obtain a new but approximate solution for the nodal displacements. A new approximate strain distribution is derived from these nodal displacements. Then the incremental constitutive model described in Chapter 2 is integrated along streamlines of constant height above the crack plane from \( x = +\infty \) to \( x = -\infty \) to obtain updated approximations for the stress, remanent strain, and remanent polarization distributions. The new remanent strain and remanent polarization distributions are then integrated on the right hand side of Equation (3.14) and the matrix solution/streamline integration procedure is repeated until a suitable level of convergence is achieved. Additional descriptions of the steady state crack growth finite element formulation can be found in Hutchinson (1974) and Landis (2003b). Once convergence is obtained, the crack tip energy release rate is computed from Equation (3.13) using the domain integral technique (Li et al., 1985).

3.3 Results

To investigate the influence of the electric field on the fracture behavior of ferroelectric materials when the electric field or the poling direction is applied parallel to
the crack front, two cases of electrical loading are considered here, the initially unpoled
and initially poled cases. After electrical loading of either case, the electric field \( E_3 \) is
kept constant and the initial state \( \varepsilon_i^0, \varepsilon_i^0 \) and \( P_3^r, P_3^r \) is attained for the fracture simulation,
after which the mechanical load is applied.

To identify important parameters that affect the toughness of ferroelectric materials, a
dimensional analysis is performed on the constitutive equations and the fundamental
differential field equations. Such analysis identifies the following normalized field
variables: \( \sigma_i/\sigma_0, \varepsilon_i/\varepsilon_c, \varepsilon_i^{r}/\varepsilon_c, D_3/P_0, P_3^r/P_0 \). All of these normalized field variables
are a function of the normalized spatial coordinates \( x_i/R \), and \( x_2/R \), and also depend
on the initial poling state \( P_3^r/P_0 \), applied electric field \( E_3^A/E_0 \), and normalized material
parameters \( E\varepsilon_c/\sigma_0, d_{33} E_0/\varepsilon_c, kE_0/P_0, \sigma_0\varepsilon_c/E_0, P_0 \), \( H_0^c/\sigma_0, H_0/E_0, d_{33}/d_{31}, \nu, \beta \) and
\( m \). Finally, the normalized steady state toughness of the material \( G_s/G_0 \) is not a
spatially varying field and hence will depend only on the initial poling state, applied
electrical loading and material parameters. It would be a rather daunting task to
investigate the effects of all ten dimensionless material quantities identified here. Instead,
this work will focus on the effects of the initial poling state \( P_3^r/P_0 \) and the applied
electrical loading \( E_3^A/E_0 \) on the toughening for a specific set of material properties.
These material properties and constitutive parameters are characteristic of a soft PLZT
material as measured by Lynch (1996), and are specifically given in Chapter 2. It should
be noted here that simply changing those parameters does not always result in a model
material that produces a reasonable constitutive response like that displayed in Figure 3.2.
Hence, if another material composition is to be modeled, it is likely that in addition to
changing the constitutive parameters listed above, the functional forms of the remanent potentials of Equations (2.20), (2.21) and (2.26) must be changed as well.

3.3.1 Switching zones

As mentioned previously, the primary result from each steady crack growth calculation is the ratio of the far field applied energy release rate, \( \mathcal{G}_{ss} \), to the crack tip energy release rate \( \mathcal{G}_{tip} \). However, prior to presenting results for the relative level of toughening, some features of the switching zones near the crack tip will be given first. Figure 3.3 illustrates the sizes and shapes of the switching zones around steadily growing cracks in initially unpoled material (a,b), and initially poled material (c). Figures 3.3d and 3.3e can be interpreted as for either initially unpoled or poled material since the constitutive response is the same for all unpoled and poled cases if the applied electric field is large enough. The specific electrical loading parameters used to generate these results are the following: unpoled (a) \( P_{3}^{c=0}/P_0 = 0 \), \( E_3^A/E_0 = 0 \), (b) \( P_{3}^{c=0}/P_0 = 0 \), \( E_3^A/E_0 = 0.5 \). (c) poled \( P_{3}^{c=0}/P_0 = 0.88 \), \( E_3^A/E_0 = 0.5 \), and (d,e) \( P_{3}^{c=0}/P_0 = 0 \) or \( P_{3}^{c=0}/P_0 \neq 0 \), \( E_3^A/E_0 = 3 \). Note that the spatial coordinates have been normalized by the length scale \( R_0 = \mathcal{G}_0 E'/3\pi\sigma_0^2 \), which can be interpreted as the size of the switching zone when the applied energy release rate is equal to \( \mathcal{G}_0 \). This normalization is used instead of \( R_0 \) in order to make the scales on each plot in Figure 3.3 comparable.

In Figures 3.3a-e the outer solid line delineates the boundary between material that is undergoing changes in remanency due to the in-plane mechanical loading and material that is not. The inner solid contour delineates the location inside the switching zone
Figure 3.3: Switching zone sizes and shapes for initially unpoled (a,b) and initially poled (c) cases. Figures (d) and (e) can be interpreted as for either initially poled or initially unpoled samples. The outermost curved contours on these plots delineate the location of the active switching boundary. The inner solid contours give the location within the active switching zones where the effective remanent strain change achieves the characteristic elastic level of $\Delta \varepsilon' = \sqrt{2} \Delta \varepsilon_0 \Delta \varepsilon'_0 / 3 = \sigma_0 / E$. The inner dashed contours give the location within the active switching zones where the change of remanent
polarization achieves the characteristic linear dielectric level $|\Delta P'_3| = \kappa E_0$. Notice that the spatial coordinates are normalized by $R_0$, which is the characteristic switching zone size when the applied energy release rate reaches $G_0$, i.e. $R_0 = G_0 E'/3\pi\sigma_0^2$. Therefore, if $G_0$ is the same for all cases, then the spatial coordinate normalizations for each plot are identical.

where the change in remanent strain reaches the characteristic elastic level of $\Delta \varepsilon' = \sigma_0/E$, and the dashed contour is where the remanent polarization change achieves the characteristic linear dielectric level $|\Delta P'_3| = \kappa E_0$. It is worth noting that in most cases the sizes of these inner switching zone contours where the effective remanent strain and polarization changes are equal to their characteristic linear values is significantly smaller than the outer switching zone boundary. This illustrates the fact that intense switching is confined to a region very close to the crack tip. Furthermore, note that the shapes of the switching zones depicted in these figures are that of the active switching zone. In other words, in the active switching zone, neighboring points at the same height above the crack plane have different remanent strain and polarization states. In contrast, in the linearly unloaded wake, neighboring points at the same height above the crack plane have identical remanent states. Lastly, material points outside of the active switching zone or the unloaded wake have a remanent state that is identical to that when the mechanical loading is initially applied.
3.3.2 The effect of initial polarization and electric field on toughening

Within this model it is assumed that crack growth occurs when $G_{\text{tip}}$ reaches the intrinsic fracture toughness of the material $G_0$. Hence the ratio $G_{ss}/G_0$ indicates the amount of toughening due to domain switching, with $G_{ss}/G_0 = 1$ corresponding to no toughness enhancement or R-curve behavior. With regard to R-curve behavior, $G_0$ should be interpreted as the applied energy release rate where crack growth first begins, and $G_{ss}$ is the steady state or plateau level of the applied energy release rate after sufficiently large amounts of crack growth. Figure 3.4 shows the ratio of $G_{ss}/G_0$ versus the applied out-of-plane electric field for a range of initial poling states. The cases associated with the solid bold curve and the dashed bold curve will be discussed first, as these cases practically envelop the others and form an inverted butterfly loop. The solid and dashed regions of the inserted hysteresis and butterfly loops in the upper left and right hand corners correspond to the solid and dashed portions of the inverted toughness butterfly loop.

To achieve the results in Figure 3.4, first consider a thermally depolarized material poled by a uniaxial out-of-plane electric field of magnitude $3E_0$. The states of electric displacement and strain for this material can be found at the upper right corners of the hysteresis loops in Figures 3.2a and 3.2b. Then, keeping this level of applied electric field fixed, steady crack growth occurs at an applied energy release rate of $G_{ss} = 1.54G_0$ and the switching zone for this case is illustrated in Figures 3.3d and 3.3e. This level of toughening is the lowest depicted on Figure 3.4. However, if the applied electric field was larger, the steady state toughness would decrease even farther, approaching $G_0$ as
$E^A \rightarrow \infty$. The reason for this behavior is that for materials with out-of-plane remanent polarization, an applied electric field in the same direction as the polarization tends to inhibit domain switching. In other words, the alignment of the polar domains with the electric field is an energetically favorable state. Since it is the domain switching process that gives rise to the dissipation of energy and the increase in fracture toughness, any phenomenon

![Graph](image)

Figure 3.4: The normalized toughness enhancement $G_{\text{sh}}/G_0$ versus the applied out-of-plane electric field for a range of initial poling states for plane-strain conditions. The bold solid line and bold dashed line on the main plot correspond to the solid and dashed lines on the outer hysteresis and butterfly loops depicted on the inserts in the upper left and right corners. The thin lines represent the toughening behavior of unpoled or partially poled materials. Points A and B are highlighted to indicate the relationships between the electromechanical constitutive response and the fracture toughness predictions.
that inhibits switching will also tend to decrease the fracture toughness. The remainder of the bold dashed curve is obtained by first poling the material with a strong electric field of \( E_s^p = 3E_0 \), then reversing the electric field to a lower or negative level of \( E_s^A \), and finally applying the in-plane mechanical loading to produce steady crack growth. During this type of initial electrical loading the butterfly behavior of the material traces out the outermost hysteresis loops depicted in Figures 3.2a and 3.2b and in the inserted plots in the upper left and right hand corners of Figure 3.4. As the electric field is removed, the inhibiting effect of the field on domain switching decreases and hence the fracture toughness increases. Furthermore, when the electric field is reversed sufficiently, it actually *enhances* the driving force for domain switching, and the fracture toughness of the material increases dramatically.

In fact, the spikes or “butterfly legs” of the toughness versus electric field curve in Figure 3.4 correspond to the legs of the butterfly loops in Figure 3.2b and the steep regions of the hysteresis loop in Figure 3.2a. However, if the reversal of the initial applied electric field is large enough, then the initial polarization of the material will be reversed as well, and the case where the polarization and electric field are aligned is revisited. Hence, as the initial electric field is driven to large negative levels, it will again inhibit domain switching and cause low values of the steady state fracture toughness. Finally, the bold solid curve is a mirror image of the bold dashed curve and is obtained by poling in the negative out-of-plane direction first. Notice that points A and B are denoted on the three loops in this figure in order to aid in the understanding of the correlation between the fundamental electromechanical constitutive behavior and the fracture toughness predictions.
Also in Figure 3.4 are the cases where the material is partially poled by a moderate electric field, then the electric field is removed, a new electric field \( E_3^A \) is applied, and finally steady crack growth proceeds due to in-plane mechanical loading. For all of the partially poled cases, negative \( E_3^A \) levels correspond very closely with the fully poled, bold dashed curve described above. The differences between the partially poled cases and the fully poled cases are evident only at intermediate positive levels of \( E_3^A \). These levels of \( E_3^A \) correspond to the linear unloading regions indicated with the arrows on Figures 3.2a and 3.2b. For high levels of \( E_3^A \) the partially poled cases eventually merge with the bold solid line that represents the situation where the material has been initially poled by a strong negative electric field. The regions of similar toughening behavior between the partially and fully poled cases can be understood by considering the hysteresis and butterfly loops of Figure 3.2a and 3.2b. Specifically, the levels of applied electric field where the toughness curves merge in Figure 3.4 coincide with the electric field levels where the linear unloading segments for the partially poled materials meet the outer hysteresis and butterfly loops in Figure 3.2. At these levels of electric field the partially poled materials show additional nonlinear behavior.

Finally, Figure 3.4 also contains a plot of toughness versus applied electric field for initially unpoled material. When considering the toughening behavior of the unpoled material during the application of electric field, it is informative to analyze the unpoled material without applied electric field. This case is presented in detail by Landis (2003b). When the material is unpoled and there are no applied electric fields, then the problem is purely mechanical and the steady state toughness of the material depends on a greatly reduced set of parameters. Dimensional analysis suggests that
\( \frac{G_{ss}}{G_0} = G(\varepsilon, E/\sigma_0, H_0^s/\sigma_0, m, n) \) for the non-electrical case. To analyze the effects of the electric field on the unpoled sample, assume that the applied electric field primarily affects only the effective switching stress \( \sigma_0 \). Specifically, it will be assumed that the switching stress can be represented as \( \sigma_0(E^s_3) \) where \( \sigma_0(E^s_3 = 0) = \sigma_0 \). Then, a first order Taylor series expansion can be applied to determine the effects of electric field on the toughness.

\[
\frac{G_{ss}}{G_0} (E^s_3) = \left. \frac{G_{ss}}{G_0} \right|_{\varepsilon_3 = 0} + \frac{\partial (G_{ss}/G_0)}{\partial (\varepsilon_3 E/\sigma_0)} \left. \varepsilon_3 E \right|_{\sigma_0 = \sigma_0} + \frac{\partial (G_{ss}/G_0)}{\partial \left( H_0^s / \sigma_0 \right)} \left. H_0^s \right|_{\sigma_0 = \sigma_0} \left( E^s_3 - E_0 \right)
\]

(3.15)

For the specific case investigated in this paper the parameters are \( \varepsilon_3 E/\sigma_0 = 3.05 \), \( H_0^s/\sigma_0 = 0.5 \), and the initial toughness and derivatives have been computed from the numerical results presented in Landis (2003b) as \( G_{ss}/G_0 \left|_{E^s_3 = 0} = 2.87 \right. \), \( \frac{\partial (G_{ss}/G_0)}{\partial (\varepsilon_3 E/\sigma_0)} \left|_{E^s_3 = 0} = 0.715 \right. \) and \( \frac{\partial (G_{ss}/G_0)}{\partial \left( H_0^s / \sigma_0 \right)} \left|_{E^s_3 = 0} = -0.5 \right. \). Finally, with the switching condition assumed in this work and given by Equation (2.17), the function \( \sigma_0(E^s_3) \) can be determined for an unpoled material as \( \sigma_0(E^s_3) = \sigma_0 \left[ 1 - (E^s_3/E_0)^2 \right] \).

Therefore, for the material properties and assumed form of the switching surface used in this work, the toughness enhancement for an unpoled material as a function of electric field can be given as

\[
\frac{G_{ss}}{G_0} (E^s_3) = 2.87 + 1.93 \left[ \frac{1}{\sqrt{1 - (E^s_3 - E_0)^2}} - 1 \right]
\]

(3.16)
Of course Equation (3.15) is a Taylor series expansion about $E^A_3 = 0$ and can be expected to be accurate only for $E^A_3$ near zero. However, a comparison of Equation (3.16) with the numerical results presented in Figure 3.4 confirms that Equation (3.16) is accurate to within 1% error for $|E^A_3| \leq 0.8E_0$, and at $|E^A_3| = 0.9E_0$ Equation (3.16) overpredicts the toughening by 6%. Finally, a similar analysis of the height of the switching zone, $h_s$, for the initially unpoled materials can be made by substituting $\bar{\sigma}_0(E^A_3)$ for $\sigma_0$ in Equation (3.7). For the assumed material properties given in this work $h_s$ is given as

$$h_s(E^A_3) = \frac{1}{3\pi} \frac{G_m E'}{\sigma_0^2} \frac{1.04}{1 - (E^A_3 - E_0)^2}$$  \hspace{1cm} (3.17)$$

For the results generated with the present model, Equation (3.17) is accurate to within 1% for $|E^A_3| \leq 0.9E_0$. Lastly, these results suggest a more general form of the toughening and switching zone heights for arbitrary switching stress dependencies as

$$\frac{G_m}{G_0}(E^A_3) = c_1 + c_2 \left[ \frac{\sigma_0}{\bar{\sigma}_0(E^A_3)} - 1 \right]$$  \hspace{1cm} (3.18)$$

and

$$h_s(E^A_3) = \frac{c_3}{3\pi} \frac{G_m E'}{\bar{\sigma}_0(E^A_3)^2}$$  \hspace{1cm} (3.19)$$

where $c_1$, $c_2$ and $c_3$ are constants.

3.3.3 Plane strain versus plane stress effects

One observation of the results displayed on Figure 3.4 is somewhat counterintuitive: If no electric field is applied, the toughness enhancement of the poled samples is actually
very slightly *smaller* than the toughness enhancement for the unpoled sample. This result is counterintuitive because one would expect that the poling process would create an out-of-plane remanent state that would allow for an increased amount of

![Graph](image)

Figure 3.5: A comparison of the effects of plane-strain versus plane-stress out-of-plane mechanical constraint on the toughness enhancement of partially poled materials. The normalized toughness enhancement $G_{ss}/G_0$ is plotted versus the initial poling field $E_3^p/E_0$. For all cases the initial poling field is removed and no subsequent electric field is applied.

dissipation from domain switching due to in-plane mechanical loads. For example, an unpoled material loaded by an in-plane tensile stress can have a maximum in-plane change in remanent strain of approximately $1.37\varepsilon_c$. In contrast, a material fully poled out-of-plane can have a maximum in-plane change of remanent strain of approximately $2.06\varepsilon_c$. Hence, it would appear that the poled material has a greater propensity for
domain switching, dissipation, and increased toughening. However, this is not the case, and the reason is the out-of-plane mechanical loading imposed by the $\sigma_{33}$ stress component arising from the plane-strain constraint. As with an applied out-of-plane electric field, domains poled out-of-plane are in a low energy state with a tensile out-of-plane stress $\sigma_{33}$. If domains aligned in the out-of-plane direction are switched towards an in-plane direction, this switching process will cause a negative out-of-plane strain. In order to enforce the plane-strain constraint, a tensile $\sigma_{33}$ stress will be induced by such a switching process. Therefore, the plane-strain constraint impedes domain switching in poled materials, and hence the full potential toughness enhancement cannot be achieved.

In order to verify and quantify the effects of the plane-strain constraint on the toughness enhancement, simulations were performed with no applied electric field on samples with differing levels of partial poling under both plane-strain and plane-stress conditions. Results for the toughness enhancement versus the level of the partial poling field are displayed in Figure 3.5. Note that the poling field of $E_3^p = E_0$ does not cause any change in remanency and so this level of poling field also corresponds to the unpoled case. Also, both of the curves in Figure 3.5 are practically flat for $E_3^p \geq 2E_0$. From this figure it is clear that the out-of-plane mechanical constraint has a significant effect on the fracture toughness. Under plane-strain conditions the toughness behavior has a very weak dependence on the level of partial poling, while for plane-stress, i.e. $\sigma_{33} = 0$, the toughness increases as the material is more fully poled. Hence, the common intuition that the toughness enhancement correlates with the potential for in-plane switching is valid for plane-stress but not for the plane-strain out-of-plane constraint.
3.3.4 The T-stress effect

Figure 3.6: The effects of the transverse stress $T_{11}$ on the toughness enhancement under plane strain conditions. The normalized toughness enhancement $\mathcal{G}_{ss}/\mathcal{G}_0$ is plotted versus the applied out-of-plane electric field with or without T-stress for (a) initially unpoled and (b) initially poled with $P_{3e}^0/P_0 = 0.88$.

The final sets of results generated in this study are displayed in Figures 3.6a and 3.6b. The plots illustrate the effects of transverse stress or T-stress on toughening behavior. To analyze the effects of T-stress, the model is modified by simply adding a constant $T_{11}$ term to the far field $\sigma_{11}$ stress given in Equation (3.8). The additional T-stress introduces a new dimensionless parameter $T_{11}/\sigma_0$ to the model. Previous models that apply a "transformation toughening" type analysis of the fracture problem (Fett et al., 2001; Cui and Yang, 2003) have predicted that a positive $T_{11}$ will decrease the level of toughening while a negative $T_{11}$ will increase the toughening for unpoled materials. The results of the present model displayed in Figure 3.6a indicate that for $E_3^d < E_0$ positive and negative T-
stress both produce very modest increases in the toughness of the unpoled material, with
the negative $T$-stress yielding a slightly larger increase. In fact, Figure 3.6a for the
unpoled material and Figure 3.6b for the poled material indicate that $T$-stress has a
significant effect on toughening only when the applied out-of-plane electric field is near
the coercive field.

3.4 Discussion

The model predicts a range of phenomena different from previous theories in that the
toughening is dependent on both the level of electric field parallel to the crack front and
on the polarization state. For poled materials, an electric field applied in the same
direction as the polarization tends to inhibit domain switching and toughening, whereas
an electric field applied opposite to the polarization directions enhances switch
toughening. For initially unpoled materials, applied electric fields below the coercive
field level enhance the fracture toughness of the material. As a complement to these
qualitative characterizations, the quantitative predictions of the model allow for a direct
comparison to some recent experimental studies.

Because it is in contrast to conventional wisdom concerning toughening due to
domain switching, the most interesting result from the model simulations is that, without
an applied electric field, the toughness of an initially unpoled material is very similar to
that of a material poled parallel to the crack front. For the material parameters
characteristic of a soft PLZT material, the model simulations have predicted that
$G_{ss} = 2.87G_0$ for the unpoled material and $G_{ss} = 2.80G_0$ for the fully poled material.
Recent experiments (Lucato et al., 2002; Hackemann and Pfeiffer 2003) also suggest that
if there is any difference between the toughening of poled and unpoled samples, then the poled specimens actually incur less toughening due to domain switching than unpoled samples. The measurements by Hackemann and Pfeiffer (2003) on a soft PZT material indicate that the toughness enhancements for both poled and unpoled samples are in the approximate range $\mathcal{G}_{ss} = 3\mathcal{G}_0 - 4\mathcal{G}_0$. Furthermore, Lucato et al. (2002) have made similar measurements on a PZT 151 composition and found that the toughness of unpoled samples is approximately $\mathcal{G}_{ss} = 2.1\mathcal{G}_0$ and that of poled samples (with short-circuited electrodes on the out-of-plane surfaces) is approximately $\mathcal{G}_{ss} = 1.9\mathcal{G}_0$. The present model prediction agrees with these experimental phenomena and shows that the out-of-plane mechanical constraint, i.e., plane-strain versus plane-stress, is the fundamental reason for this behavior.

One final set of experimental observations that warrant consideration is the study by Kolleck et al. (2000). Toughness and R-curve measurements were performed on two different PZT compositions and on a barium titanate material. In general, it was observed that the toughness of initially unpoled samples increased with increasing levels of an out-of-plane applied electric field. The same basic trend in behavior is predicted by the present model; however, a direct comparison is difficult because of the lack of nonlinear constitutive information on these specific compositions. Furthermore, if the measurements of the crack growth initiation toughness levels are accurate, the R-curve measurements on barium titanate clearly indicate that the level of initiation toughness increases with increasing electric field. Such behavior cannot be predicted from the present model, as $\mathcal{G}_0$ is an input parameter of the model. This model predicts only the multiplicative factor of toughness enhancement over the initiation toughness. In general,
the initiation toughness could depend on both the level of applied electric field and the
level of out-of-plane remanent polarization, i.e. \( G_0 = G_0(E_z, P_z') \), and this physical
behavior is required as an input to the model.

3.5 Summary

In this chapter, the constitutive model developed in Chapter 2 was simplified for the
special case where only the remanent polarization in the \( x_3 \) direction is involved. The
constitutive model was then implemented into the finite element code to study the
fracture toughening behavior of the Mode I steady crack growth with the electric field
applied parallel to the crack front. Ultimately, the present model predicts a range of
interesting effects of electric field applied parallel to the crack front on the fracture
toughness, or more specifically the toughening due to domain switching during crack
growth. The qualitative “shape” of the toughness enhancement versus applied electric
field forms an inverted butterfly loop that correlates directly with the strain versus
electric field butterfly hysteresis loop during uniaxial electrical loading. The model
predicts the counter-intuitive behavior that the fracture toughness of a material poled out-
of-plane is comparable to the toughness of an initially unpoled material. This prediction
validates and explains the previous experiments. It is demonstrated that this behavior is
primarily because of the out-of-plane mechanical constraint imposed by plane strain
conditions. In the next chapter, the effects of in-plane electric loading on the Mode-I
fracture toughening of ferroelectric ceramics will be simulated.
Chapter 4 Effects of in-plane electric fields on the toughening behavior of ferroelectric ceramics

4.1 Introduction

The constitutive model developed in Chapter 2 was successfully implemented into a finite element code in Chapter 3 to analyze the Mode-I fracture behavior of ferroelectric ceramics with electric field applied parallel to the crack front. The constitutive model is now used in this chapter to simulate the Mode-I fracture behavior of ferroelectric ceramics under combined in-plane electrical and mechanical loading.

Figure 4.1: A schematic of the in-plane electrical and in-plane mechanical loading configuration to be modeled in this work. For any given sample, the electric field and remanent polarization are aligned in \( x_1 \) or \( x_2 \) direction. The in-plane mechanical loading is simply indicative of the Mode I symmetry to be modeled in this work and should not be interpreted literally.
The scenario investigated here is illustrated in Figure 4.1. A ferroelectric material is initially poled by an electric field either perpendicular to or parallel with the direction of crack growth. After the initial poling, an electric field is applied along (positive electric field) or opposite (negative electric field) to the initial poling direction. Finally, mechanical loading is applied and crack growth occurs. A comprehensive review of the recent experimental and modeling efforts on ferroelectric fracture can be found in Zhang et al. (2001) and Chen and Lu (2002). Experimental observations of the fracture properties of ferroelectrics under the above mentioned conditions have been obtained on several materials from indentation tests and compact tension specimens. For example, using indentation tests on a PZT-8 material composition, Tobin and Pak (1993) showed that for cracks perpendicular to the poling direction, the apparent fracture toughness decreases with a positive electric field and increases with a negative field. For cracks parallel to the poling direction, their results indicated that both positive and negative electric fields have little influence on the toughening. Tobin and Pak (1993) also observed that with no applied electric field the fracture toughness was greater for cracks parallel to the poling direction than for cracks perpendicular to the poling direction.

Park and Sun (1995) investigated electric field effects on crack growth in a PZT-4 ceramic by using conventional compact tension fracture tests. Their results agree with those of Tobin and Pak (1993) for the case of electric field applied perpendicular to the crack surface. However, the Vickers indentation tests of Wang and Singh (1997) showed that if the applied electric fields are perpendicular to the crack in a PZT EC-65 ceramic, then a positive electric field impedes crack propagation, whereas a negative electric field promotes crack propagation. For the electric field parallel to the crack, their results
indicated that a negative field has little effects on the crack propagation, while a positive field impedes crack propagation. Schneider and Heyer (1999) studied the effect of a static electric field on the fracture behavior of ferroelectric barium titanate using indentation tests. Their results indicate that the measured crack length versus the applied electric field shows hysteresis similar to the strain hysteresis. In more recent compact tension tests and indentation fracture tests on a PZT-841 ceramic, Fu and Zhang (2000) observed the reduction in the fracture toughness for a positive electric field as well as for a negative electric field if the electric field is applied perpendicular to the crack surface. Lucato et al. (2002) and Hackemann and Pfeiffer (2003) performed steady crack growth experiments and recorded R-curve behavior for a variety of electrical polarization conditions. It is the existence of this R-curve behavior that indicates that the toughness variations are at least influenced by an irreversible constitutive process occurring around the crack.

Several theoretical models have been proposed in an attempt to evaluate the effect of an electric field on fracture toughness of ferroelectric ceramics. It is widely accepted that the non-linear and hysteretic constitutive behavior of ferroelectrics plays a significant role in the fracture toughness behavior. Yang and Zhu (1998) and Beom and Atluri (2003) applied transformation toughening concepts to study the effects of electric field and domain switching on the fracture toughness. Zeng and Rajapakse (2001) considered the anisotropic material properties and electromechanical coupling effect of ferroelectric ceramics and showed that a positive electric field impedes the propagation of a crack perpendicular to the poling direction while a negative field enhances it. In contrast to these approximate analytical models, the study in this chapter applies an established
constitutive model for the ferroelectric constitutive behavior within finite element computations to accurately determine the enhancement of the fracture toughness due to domain switching near a growing crack.

The contents of this chapter follow the paper of Wang and Landis (2006). The fracture model and the finite element formulation which implements the constitutive model in Chapter 2 for steady crack growth will be described first. The finite element results are then presented and analyzed. Finally, the results are discussed and compared to experimental observations.

4.2 The fracture model and finite element formulation

For the problem to be solved in this chapter, the small scale switching described in Chapter 3 will be used. The loading process also is similar to that in Chapter 3 but is entirely in-plane. Permeable electrical boundary conditions are used on the crack surfaces. The finite element formulation incorporates the constitutive model developed in Chapter 2.

4.2.1 The loading process

A schematic of the geometry and loading to be modeled here is shown in Figure 4.1. Two types of initial electrical states will be considered in this work: initially unpoled and initially poled states. The initially unpoled samples begin in the thermally depolarized state of the material. The initially poled samples are poled by applying a uniaxial electric field in the $x_\alpha$ ($\alpha = 1,2$) direction to a level of $E_\alpha^p$ and then removing the applied field. Note that the poling field $E_\alpha^p$ must be greater than the coercive field $E_0$ in order to
Figure 4.2: The uniaxial electromechanical behavior of the model material with three levels of the poling field, leading to different partially poled states. (a) The electric field versus electrical displacement hysteresis loops. (b) The electric field versus strain butterfly loop. (c) The stress versus electrical displacement depolarization loop. (d) The stress versus strain loop during depolarization. Notice that the intermediate lines in (a) and (b) represent the response during the unloading of electric field, and those in (c) and (d) represent the depolarization behavior from a partially poled state.
induce remanent polarization. This entire electrical loading procedure is performed in the absence of mechanical stress. The poling process induces both residual remanent polarization and strain in the material as referenced from the thermally depolarized state. Figure 4.2 illustrates the (a) electric displacement versus electric field, (b) strain versus electric field response during such electrical loading, (c) depolarization due to mechanical stress, and (d) the stress versus strain response during depolarization. Because of the irreversibility of the domain switching process, there is a continuous range of partial poling states that the material can attain in the range from the initially unpoled state to a fully poled state. Note that the straight lines within the loops in Figures 4.2a and 4.2b represent linear unloading during the removal of the applied electric field, and that those in 4.2c and 4.2d depict the initial behavior during depolarization by compressive stress from different partially poled states.

After the initial poling step or lack of it, the electromechanical loading history for the specimen is as follows. Electric field is applied in the $x_\alpha$ direction, again in the absence of mechanical stress. If the applied electric field is of sufficient magnitude, then poling of initially unpoled samples or a reversal of poling in initially poled samples may result. In any case, this step in the electrical loading procedure induces new states of strain and electric displacement, which will be called the initial strain $\varepsilon_{ij}^0$ and initial electric displacement $D_i^0$. The final step in the loading process is to apply the in-plane mechanical loads while keeping the applied electric field fixed at the level attained in the previous step. Under plane-strain conditions, the out-of-plane axial strain $\varepsilon_{33}$ is assumed to remain unchanged from its state after the electrical loading step, i.e. $\varepsilon_{33} = \varepsilon_{33}^0$. Steady crack growth then occurs while the in-plane mechanical loads are applied.
4.2.2 Boundary conditions

In the study of electromechanical fracture, determination of the crack face boundary conditions remains an unresolved question. Thorough reviews of the literature can be found in McMeeking (1999), Zhang et al. (2001) and Chen and Lu (2002). A point of contention among differing modeling approaches is the crack face boundary condition for the so-called "insulating" crack problem. Herein, three approaches have received considerable attention, the impermeable crack model, the permeable or "closed" crack model, and the "exact" boundary conditions. In this work, the permeable boundary condition will be used. Landis (2004d) has shown that the permeable crack boundary conditions are a reasonable approximation when the electrical discharge strength of the medium within the crack is small (the permeable conditions are exact when the discharge strength is zero).

The electrically permeable and traction free crack face boundary conditions used in the present paper are stated as

\[ D_1(r, \theta = \pi) = D_2(r, \theta = -\pi) \quad \text{and} \quad \phi(r, \theta = \pi) = \phi(r, \theta = -\pi) \quad (4.1) \]

\[ \sigma_{12}(r, \theta = \pm \pi) = 0 \quad (4.2) \]

where \( \phi \) is the electric potential, \( r \) and \( \theta \) represent a polar coordinate system centered on the crack tip, with \( \theta \) measuring the angle between the radial direction and the \( x_1 \)-axis. With the procedure similar to that outlined in Landis (2004c), the asymptotic Mode I fields for the stresses and electric potential are determined and listed in Appendix D for the piezoelectric body and applied as far-field boundary conditions. The applied energy release rate for the far field is also determined from this solution. All of these quantities
are not only dependent on $K_i$ but also dependent on the piezoelectric coefficients, Young's modulus, Poisson's ratio, and initial remanent polarization $P_{r0}^e$. Since the derivation is given in Landis (2004c) and the results for the stresses involve relatively long formulas, only the applied energy release rate, $\mathcal{G}$, is given here for brevity:

$$\mathcal{G} = \begin{cases} \frac{k_e}{4D_E} \left[ \frac{9D_E}{1 + v} + 8(2 - 3\alpha_e) \right] \frac{1 - v^2}{E} K_i^2, \text{ poled parallel to the crack plane} \\ \frac{2k_e\alpha_e}{D_E} \frac{1 - v^2}{E} K_i^2, \text{ poled perpendicular to the crack plane} \end{cases}$$  \hspace{1cm} (4.3)

where $k_e = 2E(d_{31}P_{r0}^e)^2/\rho_0^2\kappa(1-v)$, $\alpha_e = \sqrt{1/(1-k_e)}$, $D_D = k_e(1-v) + 2(\alpha_e - 1)(1+v)$, and $D_E = \kappa_e\alpha_e(1-v) + 2(\alpha_e - 1)(1+v)$. If $P_{r0}^e \to 0$, then the asymptotic fields and equation (4.3) will reduce to the solutions for the unpoled case.

### 4.2.3 Finite element formulation

To compute the electromechanical fields numerically, the following finite element formulation is applied. If we assume that the free charge density in the volume is equal to zero, the vector potential formulation proposed by Landis (2002b) can be used. The finite element formulation required to solve the steady crack growth boundary value problem is based on the variational statement

$$\int_V \sigma_{ij} \delta e_{ij} + E_i \delta D_i dV = \int_S t_i \delta u_i + \delta \phi dS$$  \hspace{1cm} (4.4)

After the implementation of the constitutive model developed in Chapter 2, equation (4.4) becomes
\[
\int_V \left[ (c_{ijkl}^{D,0} \varepsilon_{kl}^{n+1} - h_{kl}^{0} D_{k}^{n+1}) \delta e_{ij} + \left( -h_{kij}^{0} \varepsilon_{kl}^{n+1} + \beta_{ij}^{e,0} D_{l}^{n+1} \right) \delta D_{i} \right] dV = \\
\int_S (T_i \delta u_i + \phi \delta \omega) dS - \int_V \left[ (\Delta c_{ijkl}^{D} \varepsilon_{kl}^{n} - \Delta h_{kl} D_{k}^{n}) \delta e_{ij} + \left( -\Delta h_{kij} \varepsilon_{kl}^{n} + \Delta \beta_{ij}^{e} D_{l}^{n} \right) \delta D_{i} \right] dV \\
+ \int_V \left[ \left[ c_{ijkl}^{D} \varepsilon_{kl}^{e} - h_{kl} P_{k}^{e} \right] \delta e_{ij} + \left[ -h_{kij} \varepsilon_{kl}^{e} + \beta_{ij}^{e} P_{k}^{e} \right] \delta D_{i} \right] dV 
\]

(4.5)

where \( S \) is the boundary of the volume \( V \), \( \Delta \omega \) is the change of surface charge, \( c_{ijkl}^{D,0} \) and \( \Delta c_{ijkl}^{D} \) are the Cartesian components of the elastic stiffness tensor and its change due to the evolution of remanency, \( h_{kl}^{0} \) and \( \Delta h_{kl} \) are the third rank tensor of piezoelectricity and its change, \( \beta_{ij}^{e,0} \) and \( \Delta \beta_{ij}^{e} \) are the second rank dielectric tensor and its change. \( c_{ijkl}^{D} \), \( h_{kl}^{0} \), and \( \beta_{ij}^{e,0} \) are the material properties at the initial remanent state and are functions of \( P_{i}^{e,0} \) while \( \Delta c_{ijkl}^{D} \), \( \Delta h_{kl} \), and \( \Delta \beta_{ij}^{e} \) are the changes of the material properties due to the change of remanent polarization.

The tractions acting on the boundary \( S \) are given as \( t_i = \sigma_{ij} n_j \). The tractions and the electric potential (Appendix D.1~D.8) are applied on the outer boundary, and the electrically permeable boundary conditions are applied as stated in Equations (4.1) and (4.2). Returning to the finite element formulation, after the application of the appropriate finite element interpolations and the cancellation of the appropriate variational terms, the left-hand side of Equation (4.5) represents the stiffness matrix dotted with the vector of unknown nodal displacements at the \( n+1 \)th iteration. Note that this stiffness matrix depends on initial remanent polarization and this matrix remains constant for all iterations. The first integral on the right-hand side of Equation (4.5) represents the vector of known applied nodal "forces" arising from the tractions due to the mechanical loading and electrical potential due to electrical loading, which correspond to a specified level of the far field applied energy release rate. Note that these applied "forces" do not change.
from iteration to iteration. The second term on the right-hand side is an integrated body force due to the changes in the material properties. Finally, the third term on the right-hand side can be viewed as the body force due to the distributions of remanent polarization and remanent strain in the material from the $n^{th}$ iteration.

As alluded to in this discussion, the finite element Equation (4.5) is solved with an iterative technique. To begin, uniform remanent strain and polarization distributions are assumed, integrated on the right hand side of Equation (4.5), and added to the applied traction boundary conditions. Next, the system of finite element equations is solved to obtain a new but approximate solution for the nodal unknowns. A new approximate strain and electric displacement distribution is derived from these nodal unknowns. Then, the incremental constitutive model described in Chapter 2 is integrated along streamlines of constant height above the crack plane from $x = +\infty$ to $x = -\infty$ to obtain updated approximations for the stress, electric field, remanent strain, and remanent polarization distributions. The new remanent strain and remanent polarization distributions are then integrated on the right hand side of Equation (4.5), and the matrix solution/streamline integration procedure is repeated until a suitable level of convergence is achieved. Additional descriptions of the steady state crack growth finite element formulation can be found in Landis (2003b). Once convergence is obtained, the crack tip energy release rate $G_{tip}$ is computed from Equation (3.11) using an electromechanical generalization of the domain integral technique of Li et al. (1985).
4.3 Results

The goal of this work is to investigate the influence of the electric field on the fracture behavior of ferroelectric materials when the electric field or the poling direction is applied parallel or perpendicular to the crack surface in the plane of crack growth. As outlined in Section 4.2.1, two cases of electrical loading will be considered here, the initially unpoled and initially poled cases. After electrical loading of either case, the electric field \( E_\alpha, (\alpha = 1, 2) \), is kept constant and the initial state \( e^{0}_{ij}, e^{r0}_{ij}, D^0_i \) and \( P^{r0}_i \) is attained for the fracture simulation, after which the mechanical load is applied. As described in Chapter 3, it would be a sizable task to parametrically investigate the effects of all dimensionless material quantities. Instead, this work is focused on the effects of the initial remanent polarization \( P^{r0}_i / P_0 \) and the applied electric field \( E^A_i / E_0 \) on the toughening for a specific set of material properties. The material properties and constitutive parameters are used in Chapter 2 and Chapter 3 and are characteristic of a soft PLZT material as measured by Lynch (1996). Using the procedure in Section 4.2, the switching zones and variations of toughening with initial polarization and applied electric field either parallel or perpendicular to the crack surface are obtained and described below.

4.3.1 Switching zones

As mentioned in Chapter 3, the primary result of interest from each steady crack growth calculation is the ratio of the far field applied energy release rate, \( G_{ss} \), to the crack
Figure 4.3: Switching zone sizes and shapes for initially unpoled (a) and initially poled (b, c, d, e) cases. The outermost curved contours on these plots delineate the location of the active switching boundary. The inner solid contours give the location within the active switching zones where the effective remanent strain change achieves the characteristic elastic level of \( \Delta \varepsilon = \sqrt{2 \Delta \varepsilon_{\|} \Delta \varepsilon_{\perp}} / 3 = \sigma_0 / E \). The inner dashed contours
give the location within the active switching zones where the change of remanent polarization achieves the characteristic linear dielectric level \( |\Delta P| = \kappa E_0 \). Notice that the spatial coordinates are normalized by \( R_0 \), which is the characteristic switching zone size when the applied energy release rate reaches \( G_0 \), i.e. \( R_0 = G_0 E' / 3\pi \sigma_0^2 \). Therefore, if \( G_0 \) is the same for all cases, then the spatial coordinate normalizations for each plot are identical.

tip energy release rate \( G_{\text{tip}} \). However, prior to presenting results for the relative level of toughening, some features of the switching zones near the crack tip will be given first. Figure 4.3 illustrates the sizes and shapes of the switching zones around steadily growing cracks in initially unpoled material (a), and initially poled material (b,c,d,e). The specific electrical loading parameters used to generate these results are: (a) unpoled \( P_{\alpha}^r / P_0 = 0 \), \( E_\alpha^A / E_0 = 0 \), \( (\alpha = 1, 2) \), (b) poled parallel to the crack \( P_1^r / P_0 = 0.75 \) with no applied field \( E_1^A / E_0 = 0 \), (c) poled perpendicular to the crack \( P_2^r / P_0 = 0.75 \) with no applied field \( E_2^A / E_0 = 0 \), (d) poled parallel to the crack \( P_1^r / P_0 = 0.47 \) with negative applied field \( E_1^A / E_0 = -0.2 \), and (e) poled perpendicular to the crack \( P_2^r / P_0 = 0.47 \) with negative applied field \( E_2^A / E_0 = -0.2 \). Note that the spatial coordinates have been normalized by the length scale \( R_0 = G_0 E' / 3\pi \sigma_0^2 \), which can be interpreted as the size of the switching zone in a mechanically loaded unpoled material when the applied energy release rate is equal to \( G_0 \). This normalization is used instead of \( R_0 \) in order to make the scales on each plot in Figure 4.3 comparable.

In Figures 4.3a-e the outer solid black line delineates the boundary between material that is undergoing changes in remanency due to the in-plane mechanical loading and material that is not. The inner solid red contour delineates the location inside the switching zone where the change in remanent strain reaches the characteristic elastic
level of $\Delta \varepsilon' = \sigma_0 / E$, and the inner solid blue contour indicates where the remanent polarization change achieves the characteristic linear dielectric level $|\Delta P'| = \kappa E_0$. It is worth noting that in most cases the sizes of these inner switching zone contours are significantly smaller than the outer switching zone boundary. This phenomenon illustrates the fact that intense switching is confined to a region very close to the crack tip. Furthermore, note that the shapes of the switching zones depicted in these figures are those of the active switching zone. In other words, in the active switching zone, neighboring points at the same height above the crack plane have different remanent strain and polarization states. In contrast, in the linearly unloaded wake, neighboring points at the same height above the crack plane have identical remanent states. Lastly, material points outside the active switching zone or the unloaded wake have a remanent state that is identical to that when the mechanical loading is initially applied.

4.3.2 The effect of initial electrical polarization on toughening

Within this model it is assumed that crack growth occurs when $G_{\text{tip}}$ reaches the intrinsic fracture toughness of the material $G_0$. Hence the ratio $G_x / G_0$ indicates the amount of toughening due to domain switching, with $G_x / G_0 = 1$ corresponding to no toughness enhancement or R-curve behavior. With regard to R-curve behavior, $G_0$ should be interpreted as the applied energy release rate where crack growth first begins, and $G_x$, is the steady state or plateau level of the applied energy release rate. Figure 4.4 shows the ratio of $G_x / G_0$ versus the level of the initial remanent polarization under plane strain conditions. An electric field is first applied to pole the material to a given level and then removed. Thereafter, no electric field is applied. The cases for the material poled in
the \( x_3 \) direction are taken from Wang and Landis (2004) for comparison to the in-plane cases.

Prior to discussing the results for the initially poled cases, it is informative to construct a reasonable hypothesis for the qualitative behavior of the relative toughening, taking the toughening in the unpoled case as a reference. Since crack tips tend to cause higher tensile stresses in the \( x_2 \)-direction (for most polar angles around the tip) it is reasonable to assume that the materials's propensity for remanent straining in the \( x_2 \)-direction will lead to greater toughening. For example, when a material is poled by an electric field in the \( x_1 \)-direction, domains switch from being aligned closely to the \( x_2 \) and \( x_3 \) direction to the \( x_1 \)-direction. Then, when a crack tip passes through with an accompanying large \( \sigma_{yy} \) component, the domains can switch back to the \( x_2 \)-direction, causing dissipation and toughening. In contrast, a material poled by an electric field in the \( x_2 \)-direction will have most domains initially aligned closely with the \( x_2 \)-direction. When a crack tip passes, nearby domains cannot switch again towards the \( x_2 \)-axis and hence the dissipation due to domain switching will be relatively small. Applying these considerations, one would expect the following qualitative behaviors of the toughening: toughening should increase as the polarization in the \( x_1 \) or \( x_3 \)-direction increases, and toughening should decrease as the polarization in the \( x_2 \)-direction increases. However, as discussed in the previous chapter the model results show a different trend for the case in \( x_3 \)-direction.

Figure 4.4 illustrates that as the initial remanent polarization increases the toughening, \( \mathcal{G}_m/\mathcal{G}_0 \), will increase for poling parallel to the crack surface (\( x_1 \)), decrease
for poling perpendicular to the crack surface ($x_2$), and have little effect for poling parallel to the crack front ($x_3$). Hence, the qualitative hypotheses on toughening were correct for the $x_1$ and $x_2$ poling cases but incorrect for the $x_3$ case. In Chapter 3, that the relatively weak dependence of the toughening ratio on the level of poling in the $x_3$-direction was explained by considering the out-of-plane constraint imposed by the plane strain conditions. The qualitative explanation is as follows. If domains were to switch completely from being oriented towards the $x_3$-direction to an

![Graph](image)

**Figure 4.4:** A comparison of the effects of poling direction on the toughness enhancement of partially poled materials. The normalized toughness enhancement $\frac{G_{\infty}}{G_0}$ is plotted versus the initial remanent polarization state $\frac{P_i^{r,0}}{P_0}, i = 1, 2, 3$.

in-plane direction, then this would cause a large negative remanent strain component $\varepsilon_{33}^r$.

In order to maintain a total out-of-plane strain of zero, the elastic strain must then be positive and have the same magnitude as $\varepsilon_{33}^r$. This elastic strain component must arise
from an out-of-plane stress component of a magnitude approximately equal to $E\epsilon_{33}'$. Therefore, if $|\epsilon_{33}'| > \sigma_0/E$, then the out-of-plane stress will be close to $\sigma_0$ and there will be a tendency for the domains to switch back towards the out-of-plane direction. The actual events do not proceed by switching in-plane and then switching back out-of-plane, but rather by switching only a relatively small amount. Hence, the out-of-plane constraint will negate the expected toughening effect described previously. This behavior has been verified experimentally by Hackemann and Pfeiffer (2003), who observed that samples poled parallel to the crack front had practically identical toughening to unpoled samples.

### 4.3.3 Effect of applied electric field on toughening: the perpendicular poling case

In this section, the results for the case when the electric field is applied perpendicular to the crack surface (i.e. in the $x_2$-direction) will be discussed in detail. Figure 4.5 shows the ratio of $\mathcal{G}_u/\mathcal{G}_0$ versus the applied electric field in the $x_2$-direction for a range of initial poling states. The cases associated with the solid red and blue curves will be discussed first, as these cases essentially envelop the others and form an inverted butterfly loop. The red and blue regions of the inserted hysteresis and butterfly loops in the upper left and right hand corners correspond to the red and blue portions of the inverted toughness butterfly loop.

The results for fracture toughening can be explained qualitatively by considering the competing or complementary effects of the applied electric field and stress on domain switching near the crack tip. In general it is valid to assume that the crack tip stress field
will tend to elongate the material in the $x_2$-direction and will tend to cause domain switching that will produce such an elongation. First, consider a thermally depolarized material poled by a strong uniaxial electric field in the $x_2$-direction. The states of electric displacement and strain for this material can be found at the upper right corners of the hysteresis loops in Figures 4.2a and 4.2b while the field is applied. If this level of applied electric field is held fixed at $3E_0$, steady crack growth occurs at an applied

Figure 4.5: Normalized toughness enhancement $G_{sn}/G_0$ versus the applied electric field $E_2$ for a range of initial poling states for plane-strain conditions. The colored lines on the main plot are related to the corresponding colored lines on the hysteresis and butterfly loops depicted on the inserts in the upper left and right corners. Points A and B are highlighted to indicate the relationships between the electromechanical constitutive response and the fracture toughness predictions.

energy release rate of $G_{sn} = 1.4G_0$. This level of toughening is the lowest depicted on Figure 4.5. However, if the applied electric field was even greater, then the steady state
toughness would continue to decrease, approaching $G_0$ as $E_2 \to \infty$. The qualitative reason for this behavior is that both the applied electric field and the stresses near the crack tip tend to cause domain switching towards the $x_2$-direction. If the initially applied electric field is sufficiently high, then almost all of the domains that can switch towards the $x_2$-direction will have done so prior to the growth of the crack. Thereafter, due to the lack of "switchable" domains, the mechanical loads cannot cause any additional switching and it is as if the crack is running through a linear, non-dissipative, piezoelectric material. Since it is the domain switching process that gives rise to the dissipation of energy and the increase in fracture toughness, any phenomenon that inhibits any additional switching during crack growth will also tend to decrease the fracture toughness. In contrast, applied electric field and initial poling states that allow the crack tip fields to cause additional switching will enhance the fracture toughening.

The remainder of the blue portion of the curve is obtained by first poling the material with a strong electric field, then reversing the electric field to a lower or negative applied electric field level of $E_2^A$, and finally applying the in-plane mechanical loading to produce steady crack growth. During this type of initial electrical loading the electric displacement and strain behavior of the material traces out the outermost hysteresis loops depicted in Figures 4.2a and 4.2b and in the inserted plots in the upper left and right hand corners of Figure 4.5. As the electric field is removed, the inhibiting effect of the field on domain switching decreases and hence the fracture toughness increases. For positive levels of $E_2^A$, since most domains remain aligned in the $x_2$-direction, the increase in toughening remains small. However, when the applied electric field is reversed, some domains will switch towards the $x_1$-direction. Then, when the crack passes by, the crack
tip fields will switch these domains back towards the $x_2$-direction creating both dissipation and fracture toughening. The most dramatic increase in the toughening occurs very close to $E^A_2 = -E_0$, where reverse domain switching peaks due to the applied electric field. In fact, the spikes or “butterfly legs” of the toughness versus electric field curve in Figure 4.5 correspond to the legs of the butterfly loops in Figure 4.2b and the steep regions of the hysteresis loop in Figure 4.2a.

If the reversal of the initial applied electric field is large enough, i.e. for $E^A_2 < -E_0$, then the initial polarization of the material will be reversed as well, and the case where the remanent polarization and electric field are aligned is revisited. Hence, as the initial electric field is driven to large negative levels, it will again align domains in the $x_2$-direction, leaving little potential for switching due to the crack tip fields. As a result, the steady state fracture toughening is low. Finally, the red curve is a mirror image of the blue curve and is obtained by poling in the negative $x_2$-direction first. Notice that points A and B are denoted on the three loops in this figure in order to aid in the understanding of the correlation between the fundamental electromechanical constitutive behavior and the fracture toughening predictions.

Also plotted in Figure 4.5 are the cases where the material is partially poled by a moderate electric field, then the electric field is removed, a new electric field $E^A_2$ is applied, and finally steady crack growth proceeds due to in-plane mechanical loading. For all of the partially poled cases, negative $E^A_2$ levels have similar trends with the fully poled, bold dashed curve described above. The differences between the partially poled cases and the fully poled cases are evident at intermediate levels of $E^A_2$. These levels of
$E_2^A$ correspond to the linear unloading regions indicated with the arrows on Figures 4.2a and 4.2b. At the same level of $E_2^A$, the toughening decreases with increasing initial remanent polarization if $-E_0 < E_2^A < E_0$. For large levels of $E_2^A$, the partially poled cases eventually merge with the outer red and blue curves that represent the situations where the material has been initially poled by a strong electric field. The regions of similar toughening behavior between the partially and fully poled cases can be understood by considering the hysteresis and butterfly loops of Figure 4.2a and 4.2b. Specifically, the levels of applied electric field where the toughness curves merge in Figure 4.5 coincide with the electric field levels where the linear unloading segments for the partially poled materials meet the outer hysteresis and butterfly loops in Figure 4.2. At these levels of electric field, the partially poled materials commence additional nonlinear behavior.

4.3.4 Effect of applied electric field on toughening: the parallel poling case

Figure 4.6 shows the ratio of $G_{\text{int}}/G_0$ versus the applied electric field in $x_1$-direction for a range of initial poling states. Again, the cases associated with the red and blue curves will be discussed first. The red and blue regions of the inserted hysteresis and butterfly loops in the upper left and right hand corners correspond to the red and blue portions of the toughness butterfly loop. The similarities and differences between the perpendicular and parallel electrical loading cases will be explained in the following discussion.
First, consider a material poled by a strong uniaxial electric field of magnitude in the \( x_1 \)-direction. As for the perpendicular poling case, if the applied electric field is sufficiently strong, it will be able to impede domain switching such that the fracture toughening is small. The primary difference between the parallel and perpendicular cases for this strong electrical loading scenario is that for the parallel case there will always be domains available to switch towards the \( x_2 \)-direction. Then, unless the applied electric field \( E_1^A \) is extraordinarily strong, the singular crack tip fields will be able to switch domains towards the \( x_2 \)-direction creating some toughness enhancement. In fact, for an applied electric field level of \( E_0^A = 3E_0 \), the simulations predict that crack growth occurs at an applied energy release rate of \( G_{\alpha} = 2.7G_0 \) for applied field in the \( x_1 \)-direction as compared to \( G_{\alpha} = 1.4G_0 \) for applied field in the \( x_2 \)-direction. To summarize, the primary reason why toughening is low for strong electric fields applied perpendicular to the crack is that there is a dearth of domains available for switching towards the \( x_2 \)-direction. In contrast, for strong fields in the \( x_1 \)-direction, domains are available for switching, but the electric field prevents the domain switching towards the \( x_2 \)-direction. In either case, the fracture toughening decreases as the applied electric field continues to increase.

Next consider the region of the blue curve on Figure 4.6 for the region \( 0 \leq E_1^A < 3E_0 \). This region of the curve is obtained by first poling the material with a strong electric field, and then partially removing the field to a lower level of \( E_1^A \). Thereafter, the in-plane mechanical loading is applied to produce steady crack growth. As the electric field is removed, the inhibiting effects of the electric field on domain switching towards the
Figure 4.6: The normalized toughness enhancement $G_{xx}/G_0$ versus the applied electric field $E_x$ for a range of initial poling states for plane-strain conditions. The colored lines on the main plot are related to the corresponding colored lines on the hysteresis and butterfly loops depicted on the inserts in the upper left and right corners. Points A and B are highlighted to indicate the relationships between the electromechanical constitutive response and the fracture toughness predictions.

$x_2$-direction decrease and therefore the fracture toughening increases. The region of the blue toughening curve with $0 \leq E_x^A < 3E_0$ can be relatively well understood because the initial polarization state is similar for all cases within this range and only the tendency for the applied electric field to align the polarization (and hence the strain) in the $x_1$-direction needs to be considered. In contrast, a qualitative description of the behavior in the range of $-1.4E_0 < E_x^A < 0$ is considerably more difficult to construct due to the competing effects of differing potential for change in axial strain in the $x_2$-direction and
the tendency for the applied electric field to align the polarization in the $x_1$-direction. The model results for the specific material properties applied in the simulations indicate that the toughening increases in the range $-0.5E_0 < E_i^A < 0$, decreases for $-E_0 < E_i^A < -0.5E_0$, increases again for $-1.4E_0 < E_i^A < -E_0$, and finally decreases when $E_i^A < -1.4E_0$. First note that as the applied electric field traverses the range from $E_i^A = 0$ to $E_i^A = -E_0$, the remanent strain in the $x_1$-direction goes from approximately $\varepsilon'_{11} = 1.1\varepsilon_r$ to $\varepsilon'_{11} = 0$. Therefore, the case with the greatest potential for remanent straining towards the $x_2$-direction is at $E_i^A = 0$ and decreases until $E_i^A = -E_0$. Thereafter, the possible change of remanent strain towards the $x_2$-direction increases again as $E_i^A \to -\infty$. Hence, the toughening due to the potential remanent strain contribution should take the same shape as the strain-electric field butterfly loop.

The fact that the $x_1$ toughening curve does not take this shape is due to the competing effect of the applied electric field tending to align the strain in the $x_1$-direction. The strength of this competing "force" is best quantified by the energetic term $E_i^A P_i^{r,0}$. In the range from $-E_0 < E_i^A < 0$, this quantity will actually be negative suggesting that the applied electric field "helps" the strain reorient towards the $x_2$-direction. At $E_i^A = 0$ this quantity will obviously be zero and at $E_i^A = -E_0$ this quantity will also be close to zero because $P_i^{r,0} = 0$ when $E_i^A = -E_0$. For $E_i^A < -E_0$, both $E_i^A$ and $P_i^{r,0}$ will be less than zero, causing $E_i^A P_i^{r,0}$ to be positive and indicating that these levels of applied electric field inhibit the toughening. Then, when the competing effects of potential strain and applied electric field are added together, an oversimplified but qualitatively valid
understanding of the toughening curve of Figure 4.6 is obtained. Instead of the butterfly shaped toughening loop that would be expected if only the potential for remanent straining in the $x_2$-direction governed the behavior, the quantitative results predict that the effects of the applied electric field "fold" the butterfly loop "in half". Specifically, the tips of the "wings" at high applied electric field are inverted, but the "legs" remain in the same orientation.

Also shown on Figure 4.6 are the cases where the material is initially unpoled or partially poled by a moderate electric field, the electric field is removed, a new electric field $E_i^A$ is applied, and finally steady crack growth proceeds due to in-plane mechanical loading. Of special interest is the behavior of the material near zero electric field for partial poling levels of $P_i^{r,0} = 0.52P_0$ and $P_i^{r,0} = 0.7P_0$. Note that the toughening trend near zero electric field for $P_i^{r,0} = 0.52P_0$ increases with the applied electric field, whereas the trend is the opposite for a fully poled material. Also, the toughening behavior is relatively flat for the partial poling case of $P_i^{r,0} = 0.7P_0$. These intermediate poling cases illustrate the sensitivity of the toughening behavior to the initial polarization state of the material for crack growth along the applied electric field direction. Such sensitivity offers some explanation for the seemingly contradictory experimental observation for this poling case.

4.4 Discussion

The predictions of the present model are in qualitative accord with several different experimental observations. First, the model predictions displayed in Figure 4.4 indicate that the toughening is greater for crack growth parallel to the poling direction than for
crack growth perpendicular to the poling direction for the in-plane cases. This prediction is in agreement with the observations of Tobin and Pak (1993) and Lucato et al. (2002). When the polarization is parallel to the crack front, i.e. out of plane, then the model predicts that there is little to no variation in the toughness with changes in the polarization. Again, this prediction is in agreement with the experimental observations of Hackemann and Pfeiffer (2003). Additionally, the model results illustrated in Figure 4.3 indicate that domain switching is intense near the tip of the crack but diffuse towards the outer boundary of the switching zone. This prediction is also in agreement with the observations of Hackemann and Pfeiffer (2003). It should be noted that it is impossible for the transformation toughening modeling approach to capture either the out-of-plane toughening behavior or the variation of switching within the switching zone. The model predictions shown in Figure 4.5, indicating that a positive electric field reduces toughening and that a negative electric field increases toughening for polarization perpendicular to the crack, are in agreement with the observations of both Tobin and Pak (1993) and Park and Sun (1995).

As discussed above, the present model is able to explain the experimental results observed by Hackemann and Pfeiffer (2003). However, previous modeling efforts on the fracture toughness of ferroelectrics were not able to predict or explain such observations. Specifically, the previous models used the simple transformation toughening model for partially stabilized zirconia (McMeekin and Evans, 1982) to determine switching zones and fracture toughening during crack growth in ferroelectrics. These models assume that once a specific switching criterion is met, for example $E_i \Delta P'_i + \sigma_j \Delta \varepsilon'^j = G_c$ (Hwang and McMeeking, 1998), the material attains a finite transformation polarization $\Delta P'_i$ and strain
$\Delta \varepsilon'_q$ that remains fixed, i.e. frozen into the material. In contrast to the incremental flow rule used in the present model that allows both the polarization and strain to gradually evolve as the crack passes through the material, this assumption leads the transformation toughening model to predict uniform remanent strain and polarization within the switching zone and is in contrast to reality. Since intermediate remanent states are not allowed within the transformation toughening model, its prediction that the out-of-plane poled material is significantly tougher than the unpoled material is not consistent with experimental observation. Furthermore, Hackemann and Pfeiffer (2003) observed an increased concentration of switched domains close to the crack tip and a lower fraction of switched domains towards the outer boundary of the switching zone. They also measured nearly identical R-curve behavior for both unpoled and out-of-plane poled material with short-circuited electrodes. The transformation toughening models cannot capture either of these observations, while the present approach does.

4.5 Summary

The constitutive model developed in Chapter 2 was implemented into the finite element code to simulate the fracture toughening behavior of the mode-I steady crack growth with the electric field applied perpendicular or parallel to the crack surface. Similar to the out-of-plane case, the toughness enhancement versus applied electric field in the perpendicular case forms an inverted butterfly loop. However, for the parallel case, the simulation results show that the effects of the applied electric field actually "fold" the butterfly loop "in half". The competition between the mechanical switching and electrical switching driving forces is used to explain the behavior. For the same material with the
same initial poling state, under plane strain conditions and with no electric field applied, the parallel poling case shows the highest toughening, while the perpendicular poling case has less toughening than the out-of-plane case. The theoretical predictions in this chapter are also compared to experiments and demonstrate the advantage of using the current fracture model rather than the transformation toughening model in that the current model allows intermediate remanent states and is able to predict most of the experimental phenomena. In the next chapter, the mixed mode conducting fracture behavior will be analyzed.
Chapter 5 Fracture toughening behavior for conductive crack growth in ferroelectric ceramics

5.1 Introduction

The previous two chapters focused on the fracture behavior of ferroelectric materials with insulating crack face boundary conditions. However, another fracture problem of interest is the conductive crack. This chapter is devoted to the fracture behavior of ferroelectric ceramics with conductive cracks under electromechanical loadings.

![Diagram of conductive crack](image)

Figure 5.1: A schematic illustration of a conductive crack. The plus and minus signs here represent electric charges. $E_1$ is the applied electric field.

Many electromechanical devices made of ferroelectric materials use internal electrodes that can act as conductive cracks and may lead to the failure of such devices under electromechanical loading. Unlike a crack without electrical interference, the charge distributions along the upper and lower conductive crack surfaces play a great role in the propagation of a conductive crack. A schematic of a conductive crack under electrical loading is shown in Figure 5.1. When the electric field is applied parallel to the
crack surface, the electric field inside the conductive crack is zero. Therefore, the electric charges in the conductive crack will redistribute and produce an induced field that is equal to the applied one but in the opposite direction. The redistributed charges will have the same sign on the upper and lower crack surfaces. They repel each other and tend to propagate the crack. As we can see from the mechanics of a conductive crack, only the component of the applied electric field that is parallel to the crack surface can cause the crack to growth.

Among the few experimental studies, Heyer et al. (1998) developed a fracture criterion for conducting cracks in piezoelectric PZT-PIC 151 ceramics with the material poled in the direction parallel to the crack. They studied four-point bending specimens and found that an applied electric field near the coercive level, either in the positive or negative direction, actually increases the toughening. With the aid of finite element calculations without considering the nonlinear ferroelectric switching, they converted the load and electric field relationship into a fracture criterion represented by the stress intensity factor and electric field intensity factor. Also, they proposed an intrinsic fracture criterion based on the total energy release rate, but found the predictions to be quite different from the experimental fracture curve. They attributed the difference to the model's inability to account for the electromechanical coupling and domain switching in the material. The compact tension tests by Fu et al. (2000) on poled PZT-4 piezoelectric ceramics show that conductive cracks grow not only under purely mechanical loading but also under purely electrical loading. They believed that the critical energy release rates for purely mechanical and purely electrical loading are material constants with the electrical fracture toughness being much higher than the mechanical fracture toughness.
Similar results were obtained by Wang and Zhang (2001) for conductive fracture in unpoled PZT-4 ceramics. Subsequently, Zhang et al. (2003) conducted compact tension tests on unpoled PZT-4 ceramics under combined mechanical and electrical loading. With the experimental results, they proposed a fracture criterion which is an elliptic function of the normalized electric intensity factor and the normalized stress intensity factor. Zhang et al. (2004) extended the work of Zhang et al. (2003) by doing conductive fracture tests on poled PZT-8 ceramics and showing that an additional coupling term between the normalized electric intensity factor and normalized stress intensity factor should be added to the fracture criterion for the poled ceramics.

Different models have been proposed to try to explain the conductive fracture behavior of ferroelectric ceramics. Ru and Mao (1999) used a strip-saturation model of the Dugdale-type to study conducting cracks in a poled ferroelectric. They obtained the solution of near crack tip field for conducting cracks and showed that pure electric field loading applied parallel to the poling axis does not induce any stress intensity factor. Hence, their results contradict experimental observations (Heyer, 1998; Zhang et al., 2004). Rajapakse and Zeng (2001) applied the transformation toughening concepts to study the effect of combined electromechanical loading on the fracture toughness of conducting cracks. They were able to predict the experimental observations of Heyer et al. (1998) but not others. Beom and Youn (2004) also used the transformation toughening concept to examine the electrical fracture toughness under purely electrical loading by employing an idealized constitutive model for ferroelectrics. Another theoretical model on conducting cracks is the so-called charge free zone model proposed by Zhang et al. (2003) and Zhang et al. (2004). They claim that the fracture criteria derived from the
model for conductive cracks in poled or unpoled piezoelectric ceramics under electromechanical loadings agree well with the experimental results. In contrast to these approximate analytical models, this chapter applies the constitutive model established in Chapter 2 for the ferroelectric constitutive behavior within finite element computations to determine the electromechanical fields near a growing conductive crack and the enhancement of the fracture toughness due to domain switching.

Since the finite element formulation in this chapter is the same as that in Chapter 4, the remainder of this chapter is organized as follows. The fracture model for conductive cracks is introduced first. The finite element simulation for mixed-mode conductive cracks is then presented. Finally, results are discussed and compared to experiments.

5.2 Conductive boundary conditions

A schematic of the geometry and loading to be modeled here is shown in Figure 5.2. Two types of initial electrical states will be considered in this work: initially unpoled and initially poled states. The initially unpoled samples begin in the thermally depolarized state of the material. The initially poled samples are poled by applying a uniaxial electric field in the $x_1$ direction to a level of $E_1^p$ and then removing the applied field. Note that the poling field $E_1^p$ must be greater than the coercive field $E_0$ in order to induce remanent polarization. This entire electrical loading procedure is performed in the absence of mechanical stress. The poling process induces both residual remanent polarization and strain, which will be called the initial strain $e_0^0$ and initial electric displacement $D_i^0$, in the material as referenced from the thermally depolarized state.
Under plane-strain conditions, the out-of-plane axial strain $\varepsilon_{33}$ is assumed to remain unchanged from its state after the electrical loading step, i.e. $\varepsilon_{33} = \varepsilon_{33}^0$.

Figure 5.2: A schematic of the electromechanical configuration with a conductive crack to be modeled in this work. For any given sample, the electric field and remanent polarization are aligned in $x_1$ direction. The in-plane electromechanical loading is indicative of the Mode I symmetry to be modeled in this work and should not be interpreted literally.

After the initial poling step or lack of it, the electromechanical loading history for the specimen is as follows. Electric field is applied in the $x_1$ direction, again in the absence of mechanical stress. The final step in the loading process is to apply the in-plane mechanical loads while keeping the applied electric field fixed at the level attained in the previous step. Steady crack growth then occurs while the in-plane mechanical loads are applied. The applied electrical and mechanical loadings are characterized by an electric field intensity factor $K_E$ and a mode I stress intensity factor $K_I$. 
Traction free and electrically conducting crack face boundary conditions are used in
the present work, i.e.

\[ \phi(r, \theta = \pi) = \phi(r, \theta = -\pi) = 0 \rightarrow E_i = 0 \quad \text{on} \quad \theta = \pm \pi \quad (5.1) \]

and

\[ \sigma_{i2} = 0 \quad \text{on} \quad \theta = \pm \pi \quad (5.2) \]

where \( \phi \) is the electric potential, \( r \) and \( \theta \) represent a polar coordinate system centered on
the crack tip, with \( \theta \) measuring the angle between the radial direction and the \( x_1 \)-axis.

With the procedure outlined in Landis (2004c), the asymptotic Mode I and Mode E
fields for the stresses and electric potential for the poled case are determined and listed in
Appendix D for the piezoelectric body and applied as far-field boundary conditions. The
applied energy release rate for the far field is also determined from this solution. All of
these quantities are not only dependent on \( K_I \) and \( K_E \) but also on the piezoelectric
coefficients, Young's modulus, Poisson's ratio, and initial remanent polarization \( P_r^{r_0} \).
Since the derivation is given in Landis (2004c), and the results will involve relatively
long formulas, only the applied energy release rate, \( G \), for the material poled parallel to
the crack surface is given here for brevity,

\[ G = \frac{2k_e \kappa_e}{D_E} \frac{(1 - \nu^2)}{E} K_I^2 + \frac{d_{33}}{D_E} \frac{(1 - \alpha_e)(1 + \nu)}{D_E} \frac{P_r^{r_0}}{P_0} K_I K_E + \frac{D_o}{D_E} \frac{\kappa}{2} K_E^2 \quad (5.3) \]

where

\[ k_e = \sqrt{\frac{2E\left(d_{33} P_r^{r_0}\right)^2}{P_0 \kappa(1 - \nu)}}, \quad \alpha_e = \sqrt{\frac{1}{1 - k_e}}, \quad D_o = k_e(1 - \nu) + 2(\alpha_e - 1)(1 + \nu), \quad \text{and} \]

\[ D_E = k_e \alpha_e(1 - \nu) + 2(\alpha_e - 1)(1 + \nu). \] Notice that \( 0 < \frac{2k_e \kappa_e}{D_E}, \frac{D_o}{D_E} < 1 \). If \( P_r^{r_0} \rightarrow 0 \), then
\( \frac{2k_e \alpha_e}{D_e} \frac{D_\alpha}{D_e} \to 1 \) and the asymptotic field and equation (5.3) will be the solution for the unpoled case.

For the calculations to be done in this chapter, the small scale switching assumptions described in Chapter 3 will again be used. The same finite element formulation (4.5) described in Chapter 4 is used for the conductive crack which incorporates the constitutive model developed in Chapter 2. The asymptotic far field stress and electric potential (Appendix D.9–D.12) are applied as boundary conditions. With the procedures described in Section 4.2.3, the crack tip energy release rate \( G_{\text{up}} \) is computed from Equation (3.11) and results are obtained for mixed mode conductive fracture.

### 5.3 Results

The goal of this chapter is to investigate the mixed mode electromechanical fracture behavior of ferroelectric materials when the electric field or the poiling direction is applied parallel to the conducting crack surface in the plane of crack growth. As outlined in Section 5.2, two cases of electrical loading will be considered here, the initially unpoled and initially poled cases. The initial state, \( \varepsilon_{ij}^0, \varepsilon_{ij}^{\tau 0}, D_i^0 \) and \( P_i^{\tau 0} \), is attained for the fracture simulation, after which the electric field intensity factor \( K_E \) is applied and kept constant. The mechanical load is applied thereafter. As described in Chapter 3, it would be a sizable task to parametrically investigate the effects of all dimensionless material quantities. Instead, this work is focused on the effects of the initial remanent polarization \( P_i^{\tau 0}/P_0 \), the applied electric field intensity factor \( K_E \) and the applied stress intensity factor \( K_f \) on the toughening for a specific set of material properties. Here, the
material properties and constitutive parameters in Chapter 2 are again used and are characteristic of a soft PLZT material as measured by Lynch (1996). Using the procedure in Section 4.2.3 and the applied boundary conditions described in Section 5.2, different results like the switching zones, the $K$ fields and variations of toughening under combined electro-mechanical loading are obtained and described below. In all cases it is assumed that crack growth occurs when $G_{I_{0}} = G_{0}$.

5.3.1 Switching zones

As in previous chapters, some features of the switching zones near the conductive crack tip will be presented first. Figure 5.3 illustrates the sizes and shapes of the switching zones around steadily growing conductive cracks in initially unpoled material (a, b), and initially poled material (c, d). The specific loading parameters used to generate these results are: (a) unpoled, purely mechanical loading, $P_{1}^{r.0} / P_{0} = 0$, $K_{i} / K_{i}^{0} = 1.69$, $K_{e} / K_{e}^{0} = 0$. (b) unpoled, purely electrical loading, $P_{1}^{r.0} / P_{0} = 0$, $K_{e} / K_{e}^{0} = 9.68$, $K_{i} / K_{i}^{0} = 0$. (c) poled $P_{1}^{r.0} / P_{0} = 0.52$, $K_{i} / K_{i}^{0} = 2.07$, $K_{e} / K_{e}^{0} = -2.07$. (d) poled $P_{1}^{r.0} / P_{0} = 0.7$, $K_{i} / K_{i}^{0} = 2.24$, $K_{e} / K_{e}^{0} = -2.24$. Here, $K_{i}^{0}$ and $K_{e}^{0}$ are defined as $K_{i}^{0} = \sigma_{0} \sqrt{3\pi R_{0}}$ and $K_{e}^{0} = E_{0} \sqrt{3\pi R_{0}}$ where $R_{0} = \sigma_{0} E' / 3\pi \sigma_{0}^{2}$ is the characteristic switching zone size when the applied energy release rate reaches $G_{0}$ under purely mechanical loading. Again the spatial coordinates are normalized by the length scale $R_{0}$ to make the scales on each plot in Figure 5.3 comparable.
Figure 5.3: Switching zone sizes and shapes for initially unpoled (a, b) and initially poled (c, d) cases. The outermost curved contours on these plots delineate the location of the active switching boundary. The inner solid contours give the location within the active switching zones where $\Delta \mathcal{E}' = \sqrt{2 \Delta \varepsilon'_0 \Delta \varepsilon'_0/3} = \sigma_0/E$. The inner dashed contours give the location within the active switching zones where $|\Delta P'| = \kappa E_0$. Notice that the spatial coordinates are normalized by $R_0$, which is the characteristic switching zone size when the applied energy release rate reaches $G_0$ under purely mechanical loading, i.e. $R_0 = G_0 E'/3\pi\sigma_0^2$. Therefore, if $G_0$ is the same for all cases, then the spatial coordinate normalizations for each plot are identical.
In Figures 5.3a-d, we can see similar features of the switching zones as those in insulating cracks. The inner switching zone contours are significantly smaller than the outer switching zone boundary and the intense switching is confined to a region close to the crack tip. For purely mechanical loading, there is no active switching zone for polarization. While for purely electrical loading, the contour where $|\Delta P'| \geq kE_0$ contains a very large area and only a small region exists with $\Delta \bar{E}' \geq \sigma_0/E$. This phenomenon indicates that the more electrical loading comes into effect on the crack growth, the larger the polarization switching zone occupies and the higher the toughening.

5.3.2 The K fields under combined loading

The applied stress intensity factor $K_I$ normalized with $K_I^0$ is plotted against the applied electric field intensity factor $K_E$, normalized with $K_E^0$ in Figure 5.4 for the failure of ferroelectric ceramics under combined mechanical and electrical loading. As seen in the figure, under the same level of $K_I$, a larger $K_E$ is needed to cause the failure of the material with the increase of the initial poling level $P_i'/P_0$ if the electric field is applied in the opposite direction of initial poling; however, if the applied electric field is in the same direction as the initial poling, less electric field intensity factor $K_E$ is needed.
Figure 5.4: The normalized applied intensity factor during combined loadings: $\frac{K_f}{K_f^0}$ versus $\frac{K_e}{K_e^0}$.

In order to understand the toughening behavior under combined electromechanical loadings, the finite element calculation results of crack tip stress fields and electric fields are used to obtain the near-tip stress and electric field intensity factors. The crack tip stress and electric field intensity factors, $K^{tip}_f$ and $K^{tip}_e$, are different from the applied $K_f$ and $K_e$ but can be correlated as:

$$K^{tip}_f = K_f + \Delta K_f \quad (5.5)$$

$$K^{tip}_e = K_e + \Delta K_e \quad (5.6)$$
Here $\Delta K_I$ and $\Delta K_E$ characterize the change of $K_I$ and $K_E$ due to domain switching. Figure 5.5 shows the relative value of crack tip intensity factor and the applied intensity factor under combined mechanical and electrical loading. Notice that the horizontal axes in Figure 5.5a and 5.5b are different. Also note that $\frac{K_I^{up}}{K_I}$ and $\frac{K_E^{up}}{K_E}$ may be greater than 1 for the poled cases. This means switching causes “anti-shielding” of the intensity factors, but not of the energy release rate. If the electric field cannot cause crack to propagate, the change of $K_I$ increases the fracture toughness if $\Delta K_I < 0$ and the change of $K_I$ alone will reflect the fracture toughening for different situations. However, for the study in this chapter, the change of electric field intensity factor must also be considered. If the applied $K_E$ is positive, then negative $\Delta K_E$ implies that switching electrically shields the crack tip. But for negative applied $K_E$, it is positive $\Delta K_E$ that causes shielding. The toughening effect under combined electromechanical loading cannot be simply predicted from Fig. 5.5a or Fig. 5.5b.
Figure 5.5: The normalized crack tip intensity factor versus applied intensity factor during combined loadings: (a) $\frac{K_{ip}}{K_i}$ versus $\frac{K_{ip}}{K_i}$; (b) $\frac{K_{ip}}{K_i}$ versus $\frac{K_{ip}}{K_i}$. 
5.3.3 Purely mechanical loading and purely electrical loading

The first toughening behavior studied here is the initial poling effect on the toughening behavior under mechanical loading only as shown in Figure 5.6. The material is initially poled to $P_i^e$, and then the mechanical load is applied under zero electric field. From Fig. 5.6 it can be seen that the toughening increases first and then decreases with the increase of the initial poling level. This phenomenon is different from the permeable case with material initially poled in the $x_1$ direction: for the permeable case, the toughening increases with the increase of initial poling level. However, for the conducting crack, the mechanical $K_i$ loading on the poled material causes not only a crack tip stress intensity factor $K_i^{tip}$ but also a crack tip electric field intensity factor $K_e^{tip}$. The mixed-mode nature of this response makes it difficult to intuitively predict the toughening behavior.

Next, the toughening behavior for different poling levels under purely electric loading is calculated. The calculations show that for the same level of polarization, the electric field causes much higher toughening if it is applied in the opposite direction of initial poling. At the same level of $K_e$, the higher the poling level, the tougher the material, if the electric field is in the opposite direction of initial poling. However, the unpoled material shows highest toughening if the electric field is in the positive direction for the different initial poling levels. The numbers from the calculation are listed
Figure 5.6: Normalized toughness enhancement $\frac{G_n}{G_0}$ versus the ratio of initial polarization level $\frac{P_{c,0}}{P_0}$ under mechanical loading only.

here for purely electrical loading: for positive applied $K_e$, $\frac{G_n}{G_0} = 37.97$ for an unpoled material, $\frac{G_n}{G_0} = 9.22$ for $\frac{P_{c,0}}{P_0} = 0.52$, $\frac{G_n}{G_0} = 5.12$ for $\frac{P_{c,0}}{P_0} = 0.7$, and $\frac{G_n}{G_0} = 3.01$ for $\frac{P_{c,0}}{P_0} = 0.88$; for negative applied $K_e$, $\frac{G_n}{G_0} = 37.97$ for an unpoled material, $\frac{G_n}{G_0} = 68.51$ for $\frac{P_{c,0}}{P_0} = 0.52$, $\frac{G_n}{G_0} = 92.53$ for $\frac{P_{c,0}}{P_0} = 0.7$, and $\frac{G_n}{G_0} = 224.03$ for $\frac{P_{c,0}}{P_0} = 0.88$.

In Table 5.1, the ratios of the energy release rates under purely electrical loading $G_e^U$ and the energy release rate under purely mechanical loading $G^M$ from the results of the
finite element simulations and recent tests are listed. It can be seen that the calculations give a wider range of toughening behavior.

Table 5.1: The ratio of energy release rate under purely electrical loading $G^E$ and energy release rate under purely mechanical loading $G^M$ from the results of finite element simulation and recent tests.

<table>
<thead>
<tr>
<th></th>
<th>Model prediction</th>
<th>Tests results</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unpoled</td>
<td>$\frac{G^E}{G^M} = 13.6$</td>
<td>$\frac{G^E}{G^M} \approx 9$, for PZT-4 (Wang and Zhang, 2001)</td>
</tr>
<tr>
<td>Polled</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\frac{P_{r,0}}{P_0} = 0.52$</td>
<td>$\frac{G^E}{G^M} = 19.5$, negative $K_r$, $\frac{G^E}{G^M} = 2.6$, positive $K_r$</td>
<td>Initial poling level unknown $\frac{G^E}{G^M} = 25$, positive $K_r$ for PZT-4 (Ran et al., 2000)</td>
</tr>
<tr>
<td>$\frac{P_{r,0}}{P_0} = 0.7$</td>
<td>$\frac{G^E}{G^M} = 25.8$, negative $K_r$, $\frac{G^E}{G^M} = 1.4$, positive $K_r$</td>
<td></td>
</tr>
<tr>
<td>$\frac{P_{r,0}}{P_0} = 0.88$</td>
<td>$\frac{G^E}{G^M} = 85.1$, negative $K_r$, $\frac{G^E}{G^M} = 1.3$, positive $K_r$</td>
<td></td>
</tr>
</tbody>
</table>

5.3.4 Toughening behavior under mixed mode loadings

Figure 5.7 shows the toughening behavior of the material under mixed-mode electromechanical loading for different poling levels. The phenomena in figure 5.7 can again be explained by the mechanism of domain switching caused by electric field and mechanical stress. Since the mechanical loading causes domain switching towards the $x_2$ direction and the electrical loading favors switching towards the $x_1$ direction, the
mechanical stress and electric field compete with each other to cause switching towards these respective directions. For an unpoled specimen, with the increase of $K_{p}$, domains are switched towards the $x_{i}$ direction. As a result, the toughening increases with the increase of the magnitude of $K_{p}$. If the material is initially in the thermal depoled state, the opportunities to be switched to either positive $x_{i}$ or negative $x_{i}$ direction are the same. Hence, the resulted toughening effects for the same magnitude of positive or negative electric field are symmetric as shown in Figure 5.7.

For poled cases, the situation is different. For a higher initial poling level, there is a greater propensity for switching if the electric field is applied in the opposite direction of the initial poling, but there will be less opportunity for switching if the electric field is applied in the same direction of the initial poling. The mechanical loading will cause the same amount of switching no matter which direction the initial poling is in. For poled cases, the material has the highest toughening when the purely electrical loading is applied in the opposite direction of the initial poling direction. At the same level of the ratio of $\frac{K_{p}}{K_{p}^{0}}$, the toughening increases with the initial poling level if the electrical field is applied in the same direction of the poling and decreases with the initial poling level if the electrical load is applied in the opposite direction of the initial poling field.
Figure 5.7: Normalized toughness enhancement $\mathcal{G}_u/\mathcal{G}_0$ versus the ratio of $\frac{K_{1e}/K_{1e}^0}{K_1/K_1^0}$ for a range of initial poling states for plane-strain conditions. The initial poling is in the positive $x_1$ direction.

5.4 Discussion

In this section, the predictions of this study are compared to some existing experimental observations. The present simulation predicts that, for an unpoled material, the toughening under purely electrical loading is more than six times than that under purely mechanical loading. This result is in agreement with the observation of Wang and Zhang (2001). The experiments by Fu et al. (2000) predict that purely electrical loading causes much higher toughening than purely mechanical loading does for poled PZT-4 when the electric field is applied in the same direction of poling. The initial poling level in Fu et al. (2000) is likely to be low as the results in this study show that the lower the
initial poling level, the tougher the material is and the toughening is higher than that under purely mechanical loading if the electric field is applied in the direction of poling. However, if the electric field is applied in the opposite direction of initial poling, the toughening under purely electrical loading is much higher than that under purely mechanical loading.

5.5 Summary

The toughening variations for a conductive crack under combined electrical and mechanical loading are predicted for unpoled and poled ferroelectric ceramics with initial poling parallel to the crack surface. The constitutive model developed in Chapter 2 and the fracture model and finite element formulation in Chapter 4 are used. Domain switching zones around the crack tip are illustrated and compared for different poling levels. The finite element calculations also give the electromechanical fields which are used to obtain the stress and electric field intensity factor around crack tip. The results are discussed and compared to available experimental observations.
Chapter 6 Conclusions and future work

6.1 Conclusions

The model presented in this thesis differs from previous theoretical explanations of the effects of electric field and polarization on the fracture toughness of ferroelectrics in that an incremental, micro-electromechanically informed, phenomenological constitutive law has been applied instead of a discrete switching law. Additionally, in contrast to applying simplifying assumptions associated with most transformation toughening models, the details of the electromechanical fields have been obtained from finite element computations. The fields computed in this work include both the perturbing influences of ferroelectric switching and the change of the piezoelectric effect that results from such switching. The detailed constitutive model applied in this work has allowed for both qualitative and quantitative characterizations of the effects of an electric field on the toughening because of domain switching in ferroelectric ceramics. The model predicts a range of phenomena that indicate that the toughening is dependent on the level of electric field, its direction of application, and on the initial polarization state.

A multi-axial, electromechanically coupled, incremental phenomenological constitutive model for ferroelectric ceramics is first developed. The model uses the micromechanical results as the input for the thermodynamic energy functions. The flow rule and switching criteria similar to those in plasticity theory are adapted and the model is able to simulate all types of material behaviors of ferroelectric ceramics. The constitutive model is then implemented within the finite element method to study steady crack growth under electromechanical loading.
The effects of electric field and polarization in-plane or parallel to the crack front on the fracture toughening of ferroelectric ceramics are presented for an insulating crack. For fully poled materials and the initial poling in the $x_2$ or out-of-plane direction, the toughening behaves like a butterfly loop with a full cycle of electric field. An electric field applied in the same direction as the polarization tends to inhibit domain switching and toughening, whereas an electric field less than the coercive field applied opposite to the polarization directions enhances switch toughening. However, if the material is poled parallel to the crack surface, the electric field tends to fold the toughening butterfly loop in half. For initially unpoled materials, applied electric fields below the coercive field level enhance the fracture toughness of the material in either in-plane or out-of-plane directions. If the material is initially poled but there is no electric field applied during crack growth, the simulations reveal that at the same level of initial poling, the material shows the greatest toughening for initial poling in the $x_1$ direction and the lowest toughening in the $x_2$ direction. The nearly invariant toughening for different initial poling level with no electric field applied in the $x_3$ direction agrees with experimental observations and shows that the experimental results can be explained by the plane strain constraint.

This thesis also analyzed the toughening behavior of a conducting crack under combined electromechanical loadings with initial polarization or electric field applied parallel to the crack surface. Purely electrical loading causes much higher toughening than purely mechanical loading does if (a) the material is unpoled; (b) the material is poled and the electric field is applied in the opposite direction of poling; (c) the electric field is applied in the same direction of poling but the material is only slightly poled. For
polied material subject to combined electromechanical loading, the material shows highest
toughening when the electric field is applied opposite to the poling direction. The
toughening decreases with the increasing mechanical load and increases again when the
electric field applied in the direction of poling increases. The electromechanical fields
from the finite element calculations are used to generate the crack tip stress and electric
field intensity factors. In order to analyze the effect of the combined electromechanical
loading, it is not sufficient to use either crack tip stress intensity factor or electric field
intensity factor alone.

6.2 Suggestions for future research

The current model can be used to study toughening behavior of different ferroelectric
materials. Changes of material properties may change results both quantitatively and
qualitatively. For example, it is our opinion that modest changes in the shapes of the
hysteresis and butterfly loops of the material will also have a significant effect on the
shape of the $x_i$ toughening curve. Specifically, changes in the material behavior will
likely cause a shift in the location of the “fold” in the $x_i$ toughening curve.

Also the current model can be readily used to simulate the combined
electromechanical loading on the toughening behavior of a conducting crack with the
material poled perpendicular to the crack surface. Although there are no experimental
works available for this problem, a theoretical study could give guidance to experimental
researchers. Unlike the study in this thesis, this problem is not symmetric and requires a
full mesh of the crack.

The fully coupled constitutive model in this thesis can be further implemented in the
design of ferroelectric devices such as actuators, to predict their response, to assess the
device reliability, and even in control methodology. Currently we derived a consistent
tangent stiffness matrix for ferroelectric static analysis and developed a nonlinear finite
element program using the Newton’s method. The program is being tested and will be
used to analyze different ferroelectric structures.

The current constitutive is based on the isothermal assumption. However, since the
ferroelectric materials also have pyroelectric effect, a more accurate model that includes
the thermo-electro-mechanical coupling may be very useful for better understanding the
ferroelectric behaviors.
Appendix A Solving the incremental constitutive model using Newton-Raphson method

The Newton-Raphson method is widely used in root finding. Algebraically, the Newton-Raphson method derives from the familiar Taylor series expansion of a function in the neighborhood of a point,

\[ f(x + \delta) = f(x) + f'(x)\delta + \frac{f''}{2} \delta^2 + .... \]  \hspace{1cm} (A.1)

For small enough values of \( \delta \), and for well-behaved functions, the terms beyond linear are unimportant, hence \( f(x + \delta) = 0 \) implies

\[ \delta = -\frac{f(x)}{f'(x)}, \hspace{1cm} \text{or} \hspace{1cm} \delta f'(x) = -f(x). \]  \hspace{1cm} (A.2)

Newton-Raphson is not restricted to one dimension. The method readily generalizes to multiple dimensions and can be used to solve the equations in (2.17) and (2.19) for the unknowns \( \lambda \), \( \Delta \varepsilon' \), and \( \Delta P' \). The equations (2.17) and (2.19) can be rewritten as:

\[ f_i'' = \Delta P' - \lambda \frac{\partial \phi}{\partial \hat{E}_i} = 0, \]  \hspace{1cm} (A.3)

\[ f^\phi = \phi = 0, \]  \hspace{1cm} (A.4)

\[ f_y'' = \Delta \varepsilon'_y - \lambda \frac{\partial \phi}{\partial \hat{\varepsilon}_y} = 0, \]  \hspace{1cm} (A.5)

The derivatives of the functions \( f_i'' \), \( f^\phi \), and \( f_y'' \) are:

\[ \frac{\partial f_i''}{\partial \Delta P'_i} = \delta_i - \lambda \left( \frac{\partial^2 \phi}{\partial \hat{E}_i \partial \Delta P'_i} + \frac{\partial^2 \phi}{\partial \hat{E}_i \partial \hat{\sigma}_i} \frac{\partial \hat{\sigma}_i}{\partial \Delta P'_i} + \frac{\partial^2 \phi}{\partial \hat{E}_i \partial \Delta P'_i} \right), \]  \hspace{1cm} (A.6)

\[ \frac{\partial f_i''}{\partial \lambda} = -\frac{\partial \phi}{\partial \hat{E}_i}, \]  \hspace{1cm} (A.7)
\begin{align}
\frac{\partial f^r_k}{\partial \Delta \varepsilon'_i} &= -\lambda \left( \frac{\partial^2 \phi}{\partial E \partial \varepsilon'_i} \frac{\partial \hat{E}_i}{\partial \Delta \varepsilon'_i} + \frac{\partial^2 \phi}{\partial \sigma \partial \varepsilon'_i} \frac{\partial \hat{\sigma}_i}{\partial \Delta \varepsilon'_i} \right), \\
\frac{\partial f^l}{\partial \Delta P'_i} &= \frac{\partial \phi}{\partial \Delta P'_i} \frac{\partial \hat{\sigma}_i}{\partial \Delta P'_i} + \frac{\partial \phi}{\partial \hat{E}_i} \frac{\partial \hat{E}_i}{\partial \Delta P'_i}, \\
\frac{\partial f^s}{\partial \lambda} &= 0, \\
\frac{\partial f^l}{\partial \Delta \varepsilon'_i} &= \frac{\partial \phi}{\partial \Delta \varepsilon'_i} + \frac{\partial \phi}{\partial \sigma} \frac{\partial \hat{\sigma}_i}{\partial \Delta \varepsilon'_i} + \frac{\partial \phi}{\partial \hat{E}_i} \frac{\partial \hat{E}_i}{\partial \Delta \varepsilon'_i}, \\
\frac{\partial f^{er}_i}{\partial \Delta P'_i} &= -\lambda \left( \frac{\partial^3 \phi}{\partial \sigma \partial \varepsilon'_i} \frac{\partial \hat{\sigma}_i}{\partial \Delta P'_i} \frac{\partial \hat{E}_i}{\partial \Delta P'_i} + \frac{\partial^3 \phi}{\partial \hat{E}_i \partial \varepsilon'_i} \frac{\partial \hat{E}_i}{\partial \Delta P'_i} \frac{\partial \hat{E}_i}{\partial \Delta P'_i} \right), \\
\frac{\partial f^{er}_i}{\partial \lambda} &= -\frac{\partial \phi}{\partial \hat{\sigma}_i}, \\
\frac{\partial f^{er}_i}{\partial \Delta \varepsilon'_kl} &= \delta \frac{\partial \phi}{\partial \varepsilon'_kl} = -\lambda \left( \frac{\partial^2 \phi}{\partial \sigma \partial \varepsilon'_kl} \frac{\partial \hat{\sigma}_l}{\partial \Delta \varepsilon'_kl} + \frac{\partial^2 \phi}{\partial \sigma \partial \varepsilon'_kl} \frac{\partial \hat{\sigma}_l}{\partial \Delta \varepsilon'_kl} \frac{\partial \hat{\sigma}_l}{\partial \Delta \varepsilon'_kl} \right).
\end{align}

Notice that
\[
\frac{\partial}{\partial \Delta \varepsilon'_i} = \frac{\partial}{\partial \varepsilon'_i}, \quad \text{and} \quad \frac{\partial}{\partial \Delta P'_i} = \frac{\partial}{\partial P'_i}.
\]

For the function \( \phi \) used in this study, \( \frac{\partial^2 \phi}{\partial E \partial \varepsilon'_i} = 0 \), \( \frac{\partial^2 \phi}{\partial \sigma \partial \varepsilon'_i} = 0 \) and the other derivatives of \( \phi \) can also be readily obtained from (2.17). The derivatives of \( \hat{\sigma}_i \) and \( \hat{E}_i \) can be expanded from Landis (2002a) as:
\[
\frac{\partial \hat{\sigma}_l}{\partial \Delta \varepsilon'_kl} = \frac{\partial \sigma_l}{\partial \varepsilon'_kl} + \frac{\partial \sigma_l}{\partial \varepsilon'_kl} + \frac{\partial \sigma_l}{\partial P'_k}, \quad \frac{\partial \hat{E}_i}{\partial \Delta P'_i} = \frac{\partial \sigma_i}{\partial P'_i} + \frac{\partial \sigma_i}{\partial P'_i} + \frac{\partial \sigma_i}{\partial P'_i}.
\]
\[
\frac{\partial \dot{E}_i}{\partial \Delta \varepsilon_{kl}^*} = \frac{\partial E_i}{\partial \varepsilon_{kl}^*} - \frac{\partial E_i^H}{\partial \varepsilon_{kl}^*}, \quad \frac{\partial \dot{E}_i}{\partial \Delta P_j^*} = \frac{\partial E_i}{\partial P_j^*} - \frac{\partial E_i^H}{\partial P_j^*} + \frac{\partial \bar{E}_i}{\partial P_j^*}. \tag{A.17}
\]

For the material tensors used in this study, we have \( \frac{\partial \bar{\sigma}_u}{\partial \varepsilon_{kl}^*} = 0 \), \( \frac{\partial \bar{\sigma}_u}{\partial P_j^*} = 0 \), and \( \frac{\partial \bar{E}_i}{\partial P_j^*} = 0 \).

Also, \( \frac{\partial \sigma_u^H}{\partial \varepsilon_{kl}^*} = H_k^{\mu \nu} \), \( \frac{\partial \sigma_u^H}{\partial P_j^*} = H_k^{\mu \nu} \), \( \frac{\partial E_i^H}{\partial P_j^*} = H_k^{\mu \nu} \), and \( \frac{\partial \bar{E}_i}{\partial P_j^*} = U_{ij}^* \) as defined in Landis (2002a).

The derivatives of \( \sigma_u \) and \( E_i \) can be a little tricky depending on which constitutive equations are to be used. If the equations in (2.7) are used, then the derivatives of \( \sigma_u \) and \( E_i \) are all zeroes. However, if the equations in (2.9) are used, the changes of the material properties have to be considered. Using Legendre transformation and the relationship between the material tensors, the following formula are obtained:

\[
\frac{\partial \sigma_u}{\partial \varepsilon_{kl}^*} = -C_{ijkl}^{ij}, \quad \frac{\partial \sigma_u}{\partial P_j^*} = h_{ij} - C_{ijkl}^{ij} A_{kmm} + h_{ij} A_{km}^{ij}, \tag{A.18}
\]

\[
\frac{\partial E_i}{\partial \varepsilon_{kl}^*} = h_{ij}, \quad \frac{\partial E_i}{\partial P_j^*} = -\beta_i^e + h_{ij} A_{km}^{ij} - \beta_i^e A_{km}^{ij}. \tag{A.19}
\]

The definition of \( A_{km}^{ij} \) and \( A_{km}^{ij} \) can be found in Landis (2002a).

With the functions (A.3–A.5) and their Jacobians (A.6–A.14) defined, the incremental constitutive model can then be solved implementing the Newton-Raphson method.
Appendix B Fracture toughening during steady state crack growth

The applied energy release rate $\mathcal{G}$ can be used to characterize stable fracture under elastic-plastic conditions. Figure B.1 shows a typical relationship between the applied value of $\mathcal{G}$ and the amount of stable crack extension. This relationship implies ductile fracture and is known as resistance curve or R-curve. The full R-curve is a material property and is indicative of the materials toughness with toughening defined as $\frac{\mathcal{G}_m}{\mathcal{G}_0}$.

Figure B.1: Typical resistance curve (applied energy release rate $\mathcal{G}$ versus the amount of crack extension $\Delta a$).
$G_o$ is the critical value of $G$ at the onset of crack growth and can be a material property, independent of specimen geometry. In contrast, $G_\infty$, at which the steady state crack growth happens, is not a material property and can vary with specimen geometry size and the loading configuration. However, what is the source of R-curve behavior? To answer this question it is informative to consider the “flow” of energy during crack propagation.

First consider the linear elastic case with some type of separation process occurring at the crack tip. The cracked structure is loaded, and prior to crack growth work will be done by the applied loads through the displacements of the structure and elastic energy will be stored within the structure. Now, if the crack length increases by some small amount $\delta a$, the system of the applied loads will do some work $\delta W$ and the strain energy in the body will change by $\delta U$. In an elastic body where cracks are not growing we will always have $\delta U = \delta W$. However, when there is crack growth, we have

$$\delta W - \delta U = G_\Delta a$$  \hfill (B.1)

$G$ tells us how much energy is lost into the crack tip per unit area of crack advance. Here we can think of energy “flowing” into the crack tip to “feed” the separation process. Once separation is complete this energy is lost, i.e. it has been dissipated at the crack tip. These same arguments can be applied to non-linear elastic materials as well.

Next, consider what happens in an elastic-plastic material. Prior to crack advance a plastic zone will develop around the crack tip. However, as long as there is no unloading all material points close to the tip experience vary nearly proportional loading. Also, as far as the material just ahead of the tip that is almost about to separate is concerned, the amount of energy available to flow into the crack tip is still $G$. But when the crack grows, material points near the crack tip experience non-proportional loading and even
unloading. As the crack grows material points begin to load non-proportionally, or unload on a different path from which they were loaded. This process is also a dissipation process. So now for an increment of crack growth the energy balance reads

$$\delta W - \delta U = G \Delta a + \delta W_{p},$$

(B.2)

where $\delta W_{p}$ is the increment of energy dissipated by the material and usually changes with crack growth.

From (B.2), we can see that the loading system for non-elastic material has to feed energy into the dissipation due to the cohesive separation process at the crack tip, as well as the dissipation due to diffuse plastic deformation (or domain switching in this thesis) around the crack. Thus under steady state crack growth, $\frac{G_{\infty}}{G_{0}}$ is perfect to show the toughening behavior of the material since $G_{0}$ is a material constant and the higher the value of $\frac{G_{\infty}}{G_{0}}$, the more energy is used for dissipation due to plastic deformation and the tougher the material.
Appendix C Detailed finite formulation

The finite element formulation used in this thesis is based on the vector potential formulation by Landis (2002b). Since no body force and volume free charge is considered, the variational form is given as

\[ \int_V \sigma_y \delta \varepsilon_y + E \delta D_y dV = \int_S t \delta u + \phi \delta \omega dS \tag{C.1} \]

where \( \phi \) is the electric potential and \( \omega \) is the free charge per unit area residing on \( S \).

C.1 Out of plane case

For the electric components in the out of plane case, only those in \( x_3 \) are involved. The equality based on \( \delta D_y \) and \( \delta \omega \) is trivial. Based on the equality for \( \delta u \), we have

\[ \int_V \sigma_y \delta \varepsilon_y dV = \int_S t \delta u dS \tag{C.2} \]

For the material poled in \( x_1 \) direction with initial remanent strain \( \varepsilon_{10}^0 \) and initial remanent polarization \( P_{30}^r \), the first equation in (2.7) can be rewritten as

\[ \varepsilon_{10}^0 + \Delta \varepsilon_{10} - \varepsilon_{10}' = S_{1kl} \sigma_{kl} + d_{3y} E_3 \tag{C.3} \]

From \( (S_{1kl})^{-1} = C_{1kl}^{LE} \), \( \sigma_{10} \) can be solved from (C.3) as

\[ \sigma_{10} = C_{1kl}^{LE} \left( \Delta \varepsilon_{10} + \varepsilon_{10}^0 - \varepsilon_{10}' - d_{3k} E_3 \right) \tag{C.4} \]

Plug Equation (C.4) into (C.2) and after proper manipulation, the final finite element formulation (3.14) for out of plane case is obtained.
C.2 In-plane case

With an electric field or initial poling in either \( x_1 \) or \( x_2 \) direction, Equation (2.9) can be rewritten as

\[
\begin{align*}
\sigma_{ij} &= (C_{ijkl}^{(0)} + \Delta C_{ijkl}) (e_{kl}^{\varepsilon} - e_{kl}^{\varepsilon_0}) - (h_{kij}^{0} + \Delta h_{kij}) (D_{k}^{r} - P_{k}^{r}), \\
E_{i} &= -(h_{kij}^{0} + \Delta h_{kij}) (e_{kl}^{\varepsilon} - e_{kl}^{\varepsilon_0}) + (\beta_{ij}^{r,0} + \Delta \beta_{ij}^{r}) (D_{j}^{r} - P_{j}^{r})
\end{align*}
\]  
(C.6)

Notice that \( C_{ijkl}^{(0)} \), \( h_{kij}^{0} \), and \( \beta_{ij}^{r} \) are functions of \( P_{j}^{r,0} \) and \( C_{ijkl}^{(0)}, h_{kij}^{0}, \) and \( \beta_{ij}^{r,0} \) are functions of \( P_{j}^{r,0} \) with \( \Delta C_{ijkl}^{(0)} = C_{ijkl}^{(0)} - C_{ijkl}^{(0)}, \Delta h_{kij} = h_{kij} - h_{kij}^{0}, \) and \( \Delta \beta_{ij}^{r} = \beta_{ij}^{r} - \beta_{ij}^{r,0}. \) Plug Equation (C.6) into (C.1) and Equation (4.5) can be obtained.

Another kind formulation for the in-plane case is based on the increments of the variables. Equation (C.1) is now changed to

\[
\int_{V} \sigma_{ij} \delta \Delta e_{ij} + E_{i} \delta \Delta D_{i} dV = \int_{S} T \delta \Delta u_{i} + \phi \delta \Delta \omega dS
\]  
(C.7)

and Equation (C.6) changes to

\[
\begin{align*}
\sigma_{ij} &= (C_{ijkl}^{(0)} + \Delta C_{ijkl}) (e_{kl}^{\varepsilon} + \Delta e_{kl}^{\varepsilon} - e_{kl}^{\varepsilon_0} - \Delta e_{kl}^{\varepsilon_0}) - (h_{kij}^{0} + \Delta h_{kij}) (D_{k}^{0} + \Delta D_{k} - P_{k}^{r,0} - \Delta P_{k}^{r}), \\
E_{i} &= -(h_{kij}^{0} + \Delta h_{kij}) (e_{kl}^{0} + \Delta e_{kl}^{\varepsilon} - e_{kl}^{\varepsilon_0} - \Delta e_{kl}^{\varepsilon_0}) + (\beta_{ij}^{r,0} + \Delta \beta_{ij}^{r}) (D_{j}^{0} + \Delta D_{j} - P_{j}^{r,0} - \Delta P_{j}^{r}).
\end{align*}
\]  
(C.8)

Plug (C.8) into (C.7), the following formulation is obtained

\[
\begin{align*}
\int_{V} &\left[ (c_{ijkl}^{D,0} \Delta e_{kl}^{\varepsilon_0} - h_{kij}^{0} \Delta D_{k}^{r,0}) \delta \Delta e_{ij} - (h_{kij}^{0} \Delta e_{kl}^{\varepsilon_0} + \beta_{ij}^{r,0} \Delta D_{j}^{r,0}) \delta \Delta D_{i} \right] dV = \\
&\int_{S} (T \delta \Delta u_{i} + \phi \delta \Delta \omega) dS \\
&- \int_{V} \left[ (\Delta c_{ijkl}^{D} \Delta e_{kl}^{\varepsilon_0} - h_{kij}^{0} \Delta D_{k}^{r,0}) \delta \Delta e_{ij} + (\Delta h_{kij} \Delta e_{kl}^{\varepsilon_0} + \Delta \beta_{ij}^{r} \Delta D_{j}^{r,0}) \delta \Delta D_{i} \right] dV \\
&+ \int_{V} \left[ (c_{ijkl}^{D,0} + \Delta c_{ijkl}^{D}) (e_{kl}^{\varepsilon_0} + \Delta e_{kl}^{\varepsilon_0} - e_{kl}^{\varepsilon_0}) - (h_{kij}^{0} + \Delta h_{kij}) (P_{k}^{r,0} + \Delta P_{k}^{r,0} - D_{k}^{0}) \right] \delta \Delta e_{ij} \\
&+ \left[ -(h_{kij}^{0} + \Delta h_{kij}) (e_{kl}^{0} + \Delta e_{kl}^{\varepsilon_0} - e_{kl}^{\varepsilon_0}) + (\beta_{ij}^{r,0} + \Delta \beta_{ij}^{r}) (P_{k}^{r,0} + \Delta P_{k}^{r,0} - D_{k}^{0}) \right] \delta \Delta D_{i} \right] dV
\end{align*}
\]  
(C.9)
For all the formulations, appropriate formula for stiffness matrix can be obtained in Landis (2002b).
Appendix D Asymptotic fields for the permeable and conducting cracks

As mentioned in the previous chapters, the standard asymptotic fields in (3.8) are applied as far field boundary conditions for the out-of-plane cases. However, the in-plane permeable and conducting boundary conditions involve the electrical boundary conditions and the remanent polarizations. The standard asymptotic fields (3.8) no longer fit these two cases. Landis (2004c) uses complex potentials to solve for the two-dimensional, in-plane, linear piezoelectric boundary value problems and the asymptotic stress fields and electric potential are obtained for impermeable cracks and conducting cracks. For the permeable boundary conditions in this thesis, the derivation procedure is similar. Here, the asymptotic fields for the permeable and conducting cracks will be listed below. Noticed that there is no Mode II stress intensity factor $K_{II}$ or electric displacement intensity factor $K_{Ie}$ involved for the studies in this thesis.

D.1 Crack parallel to poling direction, electrically permeable

$$
\sigma_{xx} = \frac{(-a + \frac{5}{2}c - c\cos\theta + c\cos2\theta) \cos \frac{\theta}{2} - e\cos \frac{\theta}{2}}{\sqrt{r}} \cdot \frac{e\cos \frac{\theta}{2}}{\alpha_c^2 \sqrt{r_c}}, \tag{D.1}
$$

$$
\sigma_{yy} = \frac{(a + 2c) \cos \frac{\theta}{2} - c \cos \frac{5\theta}{2}}{\sqrt{r}} + \frac{e\cos \frac{\theta}{2}}{\sqrt{r_c}}, \tag{D.2}
$$
\[
\sigma_{xy} = \frac{-a \sin \frac{\theta}{2} + c \sin \frac{5\theta}{2}}{\sqrt{r}} - \frac{e \sin \frac{\theta}{2}}{\alpha_e \sqrt{r_e}}. \\
\phi = \frac{2P_0}{d_{31} \rho^* E} \left[ -4c \sqrt{r} (1 + \nu) \cos \frac{\theta}{2} + e (1 - \nu) \sqrt{r_e} \kappa_e \cos \frac{\theta_e}{2} \right].
\]

where

\[
r_e = r \sqrt{\cos^2 \theta + \left( \frac{\sin \theta}{\alpha_e} \right)^2},
\]

\[
\tan \theta_e = \frac{\alpha_e \cos \theta}{\sin \theta},
\]

\[
a = \frac{(1 - \nu) \kappa_e - 8(1 + \nu)}{4D_{ij}} \frac{K_i}{\sqrt{2\pi}},
\]

\[
c = \frac{(1 - \nu) \kappa_e}{2D_{ij}} \frac{K_i}{\sqrt{2\pi}},
\]

\[
e = \frac{2(1 + \nu) \alpha_e}{D_{ij}} \frac{K_i}{\sqrt{2\pi}}.
\]

D.2 Crack perpendicular to the poling direction, electrically permeable

\[
\sigma_{xx} = \frac{(2c - a) \cos \frac{\theta}{2} + c \cos \frac{5\theta}{2}}{\sqrt{r}} - \frac{e \alpha_e^2 \cos \frac{\theta_e}{2}}{\sqrt{r_e}}. \\
\sigma_{yy} = \frac{(2c + a) \cos \frac{\theta}{2} - c \cos \frac{5\theta}{2}}{\sqrt{r}} + \frac{e \cos \frac{\theta_e}{2}}{\sqrt{r_e}}.
\]
\[ \sigma_{xy} = \frac{-a \sin \frac{\theta}{2} + c \sin \frac{5\theta}{2} - e \alpha_\epsilon \sin \frac{\theta_\epsilon}{2}}{\sqrt{r}} - \frac{e \alpha_\epsilon \sin \frac{\theta_\epsilon}{2}}{\sqrt{r_\epsilon}}, \]  
\[ \phi = \frac{2P_0}{d_31 P' E} \left[ -4c \sqrt{r(1 + \nu)} \sin \frac{\theta}{2} - e(1 - \nu) \sqrt{r_\epsilon \kappa_\epsilon \alpha_\epsilon \sin \frac{\theta_\epsilon}{2}} \right]. \]

where

\[ a = \frac{(1 - \nu) \kappa_\epsilon \alpha_\epsilon + 8 \alpha_\epsilon (1 + \nu) K_I}{4 D_\theta \sqrt{2\pi}}, \]

\[ c = \frac{(1 - \nu) \kappa_\epsilon \alpha_\epsilon K_I}{2 D_\theta \sqrt{2\pi}}, \]

\[ e = -\frac{2(1 + \nu) \alpha_\epsilon K_I}{D_\theta \sqrt{2\pi}}. \]

D.3 Crack parallel to the poling direction, electrically conducting

\[ \sigma_{xx} = \frac{(2c - a) \cos \frac{\theta}{2} + c \cos \frac{5\theta}{2} - e \cos \frac{\theta_\epsilon}{2}}{\sqrt{r}} - \frac{e \cos \frac{\theta_\epsilon}{2}}{\alpha_\epsilon \sqrt{r_\epsilon}}, \]

\[ \sigma_{yy} = \frac{(2c + a) \cos \frac{\theta}{2} - c \cos \frac{5\theta}{2} + e \cos \frac{\theta_\epsilon}{2}}{\sqrt{r}} + \frac{e \cos \frac{\theta_\epsilon}{2}}{\sqrt{r_\epsilon}}, \]

\[ \sigma_{xy} = \frac{-a \sin \frac{\theta}{2} + c \sin \frac{5\theta}{2} - e \alpha_\epsilon \sin \frac{\theta_\epsilon}{2}}{\sqrt{r}} - \frac{e \alpha_\epsilon \sin \frac{\theta_\epsilon}{2}}{\alpha_\epsilon \sqrt{r_\epsilon}}, \]
\[ \phi = \frac{2P_0}{d_{s1}P'E} \left[ -4c\sqrt{r}(1+v)\cos\frac{\theta}{2} + e(1-v)\sqrt{r}\kappa_c\cos\frac{\theta}{2} \right], \]  
\hspace{1cm} \text{(D.12)}

where

\[ a = \frac{(1-v)\kappa_c\alpha_e - 8(1+v)}{4D_e} \frac{K_j}{\sqrt{2\pi}} + \frac{E_{d_{s1}}P'(\alpha_e + 3)}{4P_0D_e} \frac{K_j}{\sqrt{2\pi}}, \]

\[ c = \frac{(1-v)\kappa_c\alpha_e}{2D_e} \frac{K_j}{\sqrt{2\pi}} + \frac{E_{d_{s1}}P'(\alpha_e - 1)}{2P_0D_e} \frac{K_j}{\sqrt{2\pi}}, \]

\[ e = \frac{2(1+v)\alpha_e}{D_e} \frac{K_j}{\sqrt{2\pi}} - \frac{E_{d_{s1}}P'\alpha_e}{P_0D_e} \frac{K_j}{\sqrt{2\pi}}. \]
References


