PARALLEL CRACKED ITO ON PET SUBSTRATE AND ITS APPLICATION IN FREQUENCY CONTROLLED PDLC WINDOW SHUTTER

A dissertation submitted
to Kent State University in partial
fulfillment of the requirements for the
degree of Doctor of Philosophy

by
Da-Wei Lee

May, 2015
Dissertation written by

Da-Wei Lee

M.B.A., National Taiwan University, 2004
M.S., National Chiao Tung University, 1997

Approved by

____________________________________, Chair, Doctoral Dissertation Committee
Dr. John L. West

____________________________________, Member, Doctoral Dissertation Committee
Dr. Hiroshi Yokoyama

____________________________________, Member, Doctoral Dissertation Committee
Dr. Liang-Chy Chien

____________________________________, Member, Outside Discipline
Dr. Grant McGimpsey

____________________________________, Member, Graduate Faculty Representative
Dr. Brett D. Ellman

Accepted by

____________________________________, Acting Director, Chemical Physics
Dr. Hiroshi Yokoyama

____________________________________, Interim Dean, College of Arts and Sciences
Dr. James L. Blank
# TABLE OF CONTENTS

**TABLE OF CONTENTS** ........................................................................................................... iii

**LIST OF FIGURES** .................................................................................................................. vi

**LIST OF TABLES** .................................................................................................................... xiv

**ACKNOWLEDGEMENTS** ......................................................................................................... xvii

**CHAPTER 1 Introduction** ......................................................................................................... 1

1.1 Transparent Conductive Material ..................................................................................... 2

1.1.1 Importance of Transparent Conductive Material ...................................................... 2

1.1.2 Why ITO ...................................................................................................................... 3

1.1.3 Electrical and Optical Properties of ITO ................................................................. 3

1.2 Glass Substrate versus Plastic Substrate ......................................................................... 6

1.3 Cracked ITO ...................................................................................................................... 14

1.4 Heat Stabilized PET ......................................................................................................... 16

1.5 Goal and Outline ............................................................................................................... 17

**CHAPTER 2 Parallel Cracked ITO Made by Stretching Process** ......................................... 19

2.1 Initial Idea of Cracking ITO ............................................................................................ 19

2.2 Issue of Cracking ITO ..................................................................................................... 24

2.3 Solution of Cracking ITO: Stretching Process ............................................................... 42

2.4 Device: PDLC and ChLC ................................................................................................ 51

**CHAPTER 3 Parallel Cracked ITO Made by Multiple Cracking Processes** ......................... 58

3.1 Issue of Stretching Process ............................................................................................. 58

3.2 Solution of Stretching Process: Multiple Cracking Processes ..................................... 58
### 3.3 Prototype of PDLC Window Shutter having Tunable Function

**CHAPTER 4 Physics of Cracked ITO on PET Substrate**

- 4.1 Definition of Solid Mechanics
- 4.2 Previous Study of Cracks in Film bonded by Substrate
- 4.3 Cracked ITO on Different Substrates
- 4.4 Explanation of Different Cracked ITO Patterns on Different Substrates
- 4.5 How to Optimize Parallel Cracked ITO on Plastic Substrate

**CHAPTER 5 Frequency Dependent Line Width of Driving Image**

- 5.1 Frequency Independent Line Width of Conventional PDLC Device
- 5.2 Different Frequencies of Applied Voltage
- 5.3 Different Lengths of Cracked ITO Stripes
- 5.4 Response Voltage at Different Positions of Cracked ITO Stripes
  - 5.4.1 Experiment Setup for Voltage Measurement
  - 5.4.2 Measurement Result of Response Voltage

**CHAPTER 6 Analysis and Simulation**

- 6.1 Analysis of RC Circuit
- 6.2 Device Modeling
- 6.3 Measurement of Sheet Resistance
- 6.4 Measurement of Capacitance
- 6.5 LTspice Simulation
- 6.6 Analytical Solution of Cracked ITO Device

**CHAPTER 7 Summary**
7.1 Dissertation Contribution................................................................. 148
7.2 Future Study Topics of Cracked ITO................................................ 149

REFERENCES........................................................................................................ 151
LIST OF FIGURES

Figure 1.1: Structure and components of TFT LCD [8]........................................ 2
Figure 1.2: Current and forecast market share of different transparent conductive materials [12]........................................................................................................ 3
Figure 1.3: Optimization on both the electrical and optical properties of ITO by adjusting the weight percentage of tin oxide [10]. ................................................. 5
Figure 1.4: Transmission and reflection spectrum of thickness 200 nm ITO films treated by different annealing temperatures [16]. ........................................... 6
Figure 1.5: In May 2010, Sony Corporation demonstrated 80 µm thickness 4.1 inch wide full color OTFT-driven OLED flexible display made by 20 µm flexible substrates, which can be wrapped around by a cylinder with 4 mm radius [24]...... 7
Figure 1.6: The working principle of PDLC [35]....................................................... 9
Figure 1.7: Yang [43], (a) Cholesteric Liquid Crystal (ChLC) device structure and two stable optical states while no electric field is applied between two layers of ITO. (b) The bistable behavior of ChLC: reflection versus applied voltage V, the reflection is measured after applying V between two layers of ITO for several seconds and then turned off. There is no voltage applied on CHLC layer while the reflection is measuring......................................................... 12
Figure 1.8: The working principle of Boogie Board™, on which the finger tip or stylus releases pressure on the surface and deform the plastic film which is elastic, therefore the cholesteric liquid crystal underneath is changed into planar state [48]. .................................................................................................................. 14
Figure 2.1: Long-lasting parallel cracks appeared near the cutting edge of PEDOT on ITO or PET substrate. (a) (c) (e) are SEM pictures of “PEDOT on ITO on PET” sample with magnification of 500, 1000, and 2500 respectively. (b) (d) (f) are SEM pictures of “PEDOT on PET” sample with magnification of 500, 1000, and 2500 respectively................................................................. 20
Figure 2.2: The first try of making parallel ITO cracks on PET substrate. A piece of 2 cm × 8 cm ITO on PET film (a) is manually attached and moved at a rectangle corner of a table (b) with ITO side on top. The resulting cracks in ITO are observed under optical microscope, as shown in (c) to (f). .......................... 23

Figure 2.3: The second try of making parallel ITO cracks on PET substrate. (a) A manually bending apparatus designed to make parallel cracked ITO stripes on PET substrate. Sphere beads of diameter 4mm are distributed between two glass plates to maintain a gap of 4 mm. Top plate is manually moved with a speed of 1 cm / sec, while the bottom plate is fixed. (b) The real apparatus designed in (a). (c) The resulting cracked ITO on PET substrate, which is pulled to be straight. (d) The resulting cracked ITO on PET substrate, which is free standing as a roll of plastic film........................................................................................................ 26

Figure 2.4: The third try of making parallel ITO cracks on PET substrate. (a) The cracked ITO on PET attached on the curved surface of 3 cm diameter cylinder. (b) The curved cracked ITO on PET substrate is immersed in the 2.5 wt % HCl in H₂O solution........................................................................................................ 29

Figure 2.5: The fourth try of making parallel ITO cracks on PET substrate. (a) Two pieces of glass substrate and cracked ITO sample are adhered by using 5 minutes epoxy and four clips. (b) Two multi-meters are used to measure the resistance of R_AB and R_CD, while cracked ITO sample is heated. (c) Cracked ITO sampled is heated while loading 429 grams metal block........................................... 33

Figure 2.6: Gohil [110], Schematic representation of PET fiber model (a), and structure situation in the various temperature regimines for the oriented PET film (b), L represents long spacings......................................................... 37

Figure 2.7: An apparatus consisted of two parallel rails which provide precise and reproducible rolling process of making parallel cracked ITO on PET substrate. (a) Side view. (b) Top view................................................................. 43

Figure 2.8: Cracks were formed by the bending induced stress, which is the root cause of R_perp/R_para................................................................. 45
Figure 2.9: Intersection between two neighboring cracks can occur, if the top plate is not precisely shear-stressed along two parallel rails. .................................................. 46
Figure 2.10: Uniaxially cracked ITO prepared by extension and compression stress. .................................................................................................................................. 47
Figure 2.11: Biaxially cracked ITO squared islands prepared by extension stress, bending the ITO on PET film along two orthogonal directions. ................................. 47
Figure 2.12: Contact between ITO stripes leads to spreading of an applied electric field. .................................................................................................................................. 48
Figure 2.13: Uniaxially stretching system in Professor Mukerrem Cakmak research group in the University of Akron. ........................................................................................................... 49
Figure 2.14: After uniaxially stretching, the separation between cracked ITO stripes is increased to 50 nm. .................................................................................................................. 49
Figure 2.15: For the uniaxially stretching process, dumbbell shape ITO on PET film is designed to reduce the compression in the middle region. ........................................ 50
Figure 2.16: For the uniaxially stretching process, rectangle shape ITO on PET film produces compression cracks perpendicular to parallel cracks the stretching is meant to enhance........................................................................................................... 50
Figure 2.17: The structure (a) and the real device (b) of a small area (3 cm × 3 cm) PDLC device made by a cracked ITO on bottom PET substrate and a continuous ITO on top PET substrate. (b-1) Two wider 5mm lines addressed by 20k Hz and 40 volts sine wave. (b-2) Two thinner 1mm lines addressed by 200k Hz and 40 volts sine wave .................................................................................................................................. 53
Figure 2.18: The structure (a) and real device (b) of display area 3 cm × 3 cm, 7 × 7 pixels, pixel size 2 mm × 2 mm, PM ChLC display in which two pieces of cracked ITO on PET substrate are orthogonally assembled. The frequency of applied voltage is 500 Hz, the amplitude of applied voltage is 80 volts, and the cell gap is 15 microns. .................................................................................................................................. 56
Figure 3.1: (a) Comparison of cracking ITO processes, (a, left) Previous method, first the ITO PET film is outwardly bent while ITO layer is on the outside, and then
it is stretched, (a, right) Improved method, first the ITO PET film is outwardly bent while ITO layer is on the outside, and then it is inwardly bent while ITO layer is on the inside. (b) Microscopic images of improved cracked ITO on PET substrate, (b, left) After the first outwardly bending process, (b, right) After the inwardly bending process.

Figure 3.2: A real window size (26 cm × 42 cm) PDLC window. (a) Off state. (b) On state, applied with 60 volts 50 Hz for the whole display area. (c) The contact electrodes from driving system are attached on cracked ITO stripes.

Figure 3.3: The “window curtain” function of PDLC window. (a) Drawing to reveal the bottom area of the window. (b) Drawing to reveal the middle and bottom areas of the window. 60 volts 50 Hz external voltage is applied on selected electrodes in the transparent area.

Figure 3.4: The “window shutter” or “venetian blind” function of PDLC window. (a) Tuning to reveal wider 3 cm transparent lines, 60 volts 2000 Hz voltage is applied on all electrodes. (b) Tuning to reveal narrower 3 mm transparent lines, 60 volts 5000 Hz is applied on all electrodes.

Figure 3.5: The curtain and shutter functions of PDLC window. (a)~ (b) Drawing to reveal the bottom area (a) or middle and bottom areas (b) of the window, and tuning to reveal wider 3 cm transparent lines, 60 volts 2000 Hz voltage is applied on selected electrodes. (c) ~ (d) Drawing to reveal the bottom area (c) or middle and bottom areas (d) of the window, and tuning to reveal narrower 3 mm transparent lines, 60 volts 5000 Hz voltage is applied on selected electrodes.

Figure 4.1: Stolfi [67], a picture to illustrate the definition of stress. The force applied by a body (top sphere) on another body (bottom sphere) through the connection surface (yellow disk) that separates them is represented by the large purple arrow. The stress (the smaller arrows distributed on the yellow disk) is the force divided by the area of the connection surface.

Figure 4.2: eFunda Inc. [72], Definition of stress (a) and stress tensor (b).
Figure 4.3: Mircalla22 [74], An example of calculating the strain of the deformation of a rectangle in x y plane. .................................................................................................................. 74

Figure 4.4: Batzle [77], the definition of vertical strain, Young’s modulus, lateral strain, and Poisson’s ratio. .................................................................................................................. 75

Figure 4.5: The definition of shear stress, shear strain, and shear modulus. ............. 76

Figure 4.6: Beuth [63], an isotropic film (labeled 1) having thickness h is bonded on an isotropic substrate (labeled 2), when stress $\sigma$ is applied on the film, the steady state of cracked film will be: (a) fully cracked film, in which a single crack extending down to the film/substrate interface is generated, or (b) partially cracked film, in which a single crack extending down to the depth of $a$ in the film is generated. (c) Perspective picture of (a). .......................................................................................... 77

Figure 4.7: Xia [64], for $\beta = 0$ (the dependence on $\beta$ is weak), the reference length $\ell$ which characterize the exponential decay of the changes transverses to a steady-state fully cracked crack in a film. The normalized length $\ell / h$ which defines the in-plane resistance of the film is determined by the function $g(\alpha, \beta)$. ........................................80

Figure 4.8: Xia [64]. Energy release rate at each crack tip for steady-state channeling of parallel cracks in the film bonded on substrate. The upper curve applies to the simultaneous advance of all cracks, and the lower curve applies to the sequential process where a new set of cracks propagate midway between a previously formed set of cracks. .......................................................................................... 89

Figure 5.1: The PDLC window in which the bottom plate is made by conventional patterned ITO stripes on PET substrate, and the top palte is made by continuous ITO plane on PET substrate. Two neighboring ITO stripes in the bottom plate are completely isolated by using the photo-lithography or laser patterning process. The line width of driving image is independent on the frequency of applied voltage, and it is only dependent on how many connections are built between the ITO stripes and the electrodes. .......................................................................................... 93

Figure 5.2: The PDLC window in which the bottom plate is made by our cracked ITO stripes on PET substrate, and the top palte is made by continuous ITO plane on
PET substrate. Two neighboring ITO stripes in the bottom plate are still connected. The line width of driving image is dependent on the frequency of applied voltage. 94

Figure 5.3: The line width of driving image is dependent on the frequency of applied voltage and the length of cracked ITO stripes. (a) (c) (e): 5000 Hz and 60 volts voltage is applied on the 2 mm adhesive copper tape. (b) (d) (f): 500 Hz and 60 volts voltage is applied on the 2 mm adhesive copper tape. 98

Figure 5.4: The system and device of measuring the amplitude and phase of response voltage at different positions on the cracked ITO stripes. (a) The illustration of the measurement system including a lock-in amplifier and a voltage follower, and the top view of device. (b) The side view of device. (c) The picture of measurement system and device. (d) The picture of measuring the device by using a probe from the lock-in amplifier. 105

Figure 5.5: The plots of amplitude and phase of response voltages at different positions of cracked ITO on PET substrate, while the frequency of applied voltage is ranging from 1 k Hz to 125 k Hz. These plots are transferred from Table 5.3 by dividing a factor to make the amplitude at 0 cm to be 100%, and subtracting a factor to make the phase at 0 cm to be 0. 109

Figure 6.1: Two serially connected resistors driven by an AC voltage. 110

Figure 6.2: Two serially connected capacitors driven by an AC voltage. 111

Figure 6.3: A resistor serially connected with a capacitor driven by an AC voltage. 112

Figure 6.4: The quantitative relationship between $\frac{I}{V}$ and $\frac{\omega}{\frac{1}{R \cdot C}}$ in the circuit of a resistor $R$ serially connected with a capacitor $C$. $I$ is the amplitude of response current, $V$ is the amplitude of applying AC voltage, $\omega$ is the frequency of applying AC voltage. 115

Figure 6.5: The “Cause and Effect” relationship explaining the frequency dependent line width of driving image in the PDLC window made by parallel cracked ITO on PET substrate. 119
Figure 6.6: The structure of PDLC window made by our parallel cracked ITO stripes on PET substrate. (a) In the assembling process, the cracked ITO substrate is flipped over to face the plane ITO substrate. (b) The side view of PDLC window along the Y direction, the dimensions of cracks, cracked ITO stripes, and each layer are illustrated. (c) The layout of adhesive copper tape on top substrate and the tiny connections between neighboring parallel cracked ITO stripes. The tiny and uniform connections shown in Figure 6.6 (b) and (c) are representative symbols of the basically unknown structure by which two neighboring cracked ITO stripes are connected. .......................................................... 122

Figure 6.7: The device modeling (i.e. equivalent circuit) of PDLC window made by cracked ITO on PET substrate. .......................................................... 124

Figure 6.8: The line contact electrodes (a), and squared film resistors (b) (c), are used to measure the sheet resistance of ITO film on PET substrate. .......... 128

Figure 6.9: (a) The dimensions of cracked ITO stripes and cracks. (b) The definition of sheet resistance $R_{\text{para}}$ and $R_{\text{perp}}$, where $R_{\text{para}}$ is parallel and $R_{\text{perp}}$ is perpendicular to the direction of ITO strips. .......................................................... 129

Figure 6.10: The definition of $R_{\text{ITO,para}}$ (a), $R_{\text{ITO,perp}}$ (b), $R_{\text{crack,para}}$ (c), $R_{\text{crack,perp}}$ (d), and how to calculate their values by using the measured sheet resistance of ITO or crack (e) ........................................................................................................ 132

Figure 6.11: The value of capacitance is determined by the structure and dimensions of a capacitor. .......................................................... 137

Figure 6.12: The equivalent circuit of PDLC window made by cracked ITO on PET substrate, in which the length of cracks is 10 cm. .......................................................... 138

Figure 6.13: (a) The equivalent circuit consisting of 200 resistors of 150 ohm and 200 capacitors of 6.3 pF. (b) An enlarged picture for the circuit in red line region of (a). (c) The simulation result of this equivalent circuit by using LTspice: the green line is the applied voltage 60 volts 5000 Hz sine wave, the dark blue line is the response voltage of the capacitor of 1260 pF, the red line is the response voltage of
the 100\textsuperscript{th} capacitor of 6.3 pF, and the light blue line is the response voltage of the 200\textsuperscript{th} capacitor of 6.3 pF.

Figure 6.14: The equivalent circuit consisting of 1000 resistors of 150 ohm and 1000 capacitors of 6.3 pF. The green line is the applied voltage 60 volts 5000 Hz sine wave, the dark blue line is the response voltage of the capacitor of 1260 pF, the red line is the response voltage of the 200\textsuperscript{th} capacitor of 6.3 pF, the light blue line is the response voltage of the 400\textsuperscript{th} capacitor of 6.3 pF, the pink line is the response voltage of the 600\textsuperscript{th} capacitor of 6.3 pF, the gray line is the response voltage of the 800\textsuperscript{th} capacitor of 6.3 pF, and the dark green line is the response voltage of the 1000\textsuperscript{th} capacitor of 6.3 pF.

Figure 6.15: Edgar [97], (a) Circuit element, and (b) Symmetrical circuit of lumped-element transmission line.

Figure 6.16: The equivalent circuit of cracked ITO PDLC device, which is a ladder array of this primary building block.
LIST OF TABLES

Table 1.1: Photon energy calculated in near infrared, visible, and ultraviolet regions. Frequency = light speed \((3 \times 10^8 \text{ m/sec}) / \text{wavelength}\). Photon energy = Planck constant \(h (4 \times 10^{-15} \text{ eV \cdot sec}) \times \text{frequency}\). ................................................................. 4

Table 1.2: Comparing glass substrate with plastic film in the PDLC window applications..............................................................................................................................11

Table 2.1: The perpendicular resistance \(R_{AB}\) and the parallel resistance \(R_{CD}\) are measured by using multi-meter on ITO on PET film as shown in Figure 2.2 (a). .... 23

Table 2.2: The measurement of resistance \(R_{AB}\) and \(R_{CD}\) on flat surface and curved surface, before and after cracking ITO on PET substrate by using a manually bending apparatus with tight radius 2 mm of bending curvature. ......................... 27

Table 2.3: The measurement of resistance \(R_{AB}\) and \(R_{CD}\) after the process of “Wet etching of cracked ITO on PET substrate” in which PET substrate is bent as a curved sample........................................................................................................................................ 30

Table 2.4: The measurement of resistance \(R_{AB}\) and \(R_{CD}\) in the process of “Heating and Loading of cracked ITO on PET substrate” ......................................................... 35

Table 2.5: The comparison of four methods tried to increase the value of \(\frac{R_{AB}}{R_{CD}}\).

Method 1: manually stretching two sides of ITO on PET substrate. Method 2: the ITO on PET substrate is bent as a curvature of tight radius, and two glass plates are manually rolled to make parallel ITO cracks on PET substrate. Method 3: after making parallel ITO cracks by using method 2, the sample is immersed into 2.5 wt % HCl and etched for a period of time. Method 4: after making parallel ITO cracks by using method 2, the sample is heated in a heat oven and hung by a 429 gram metal block........................................................................................................................................ 41
Table 2.6: Different values of $R_{perp}/R_{para}$ were caused by different radiiuses of bending curvature (i.e. different separations between two plates). Corresponding OM and SEM pictures were also included. ...................................................... 45

Table 4.1: The comparisons of four samples of ITO on plastic substrate, the resistance ration after rolling process and stretching process are summarized. ................................. 81

Table 4.2: The comparison of microscopic pictures (1000×) for three Genvac samples after rolling process and stretching process is summarized. .............................. 83

Table 4.3: The comparison of microscopic pictures (400×, 200×, 100×) for three Genvac samples after stretching process is summarized. ........................................... 84

Table 4.4: The looked up values of Young’s modulus, Poisson ratio, and the calculated values of Shear modulus: ITO, PET, PC, and PEN. ............................................. 85

Table 4.5: $E, E_s, \alpha, \beta$ in three systems of “film bonded on substrate”: ITO film on PET substrate, ITO film on PC substrate, and ITO film on PEN substrate. ............. 85

Table 4.6: Beuth [63], the look-up table of $f(\alpha, \beta)$ and $g(\alpha, \beta)$ in fully cracked film................................................................. 87

Table 4.7: $f(\alpha, \beta)$ and $g(\alpha, \beta)$ are calculated by using linear interpolation and the values within the red frames in Table 4.6. ..................................................... 87

Table 4.8: Four samples of parallel cracked ITO on PET substrate having different thickness of ITO film and PET substrate are outwardly bent 6 times at 3 mm radius of curvature. ............................................................... 90

Table 5.1: The comparison of the frequency dependent line width of driving image on PDLC samples made by different lengths of cracked ITO stripes. The width of adhesive copper tape is 2 mm. The amplitude of the applied voltage is 60 volts. .... 99

Table 5.2: The tuning ratio and tunability (i.e. values in the row of 50 Hz), which provide quantitative analysis for the frequency dependent line width of driving image. Three PDLC samples with different lengths of cracked ITO stripe are compared. This table is converted from Table 5.1. ......................................................... 101
Table 5.3: The amplitude and phase measurement results of response voltages at different positions of cracked ITO on PET substrate, while the frequency of applied voltage is ranging from 1 k Hz to 125 k Hz.

Table 6.1: The measured sheet resistances, $R_{\text{para}}$ and $R_{\text{perp}}$, by using different lengths of line contact electrodes and different sizes of cracked ITO squares.

Table 6.2: The capacitance measurement of PDLC samples, and the calculation of PDLC capacitance corresponding to a cracked ITO stripe in our model.
ACKNOWLEDGEMENTS

I would like to acknowledge many people that have helped me on my path to this degree. I will start with my advisor, Professor John L. West. His amazing insight into the unusual possibility from a common experimental result was the origin of everything found in this thesis. Without his guidance, I would have ended this study three years ago without having any valuable result. And not only has he been a supervisor, but also a partner literally. We started up a company Flexible ITO Solutions (FITOS) LLC in 2013 to commercialize our unique technology. I have enjoyed this exciting journey and learned a lot of precious experiences, all of these are offered from Professor West.

I would also like to thank the valuable co-advisors with whom I have worked along the way: Professor Hiroshi Yokoyama, for the basic physics study of cracks behavior in Chapter 4, his energetic directions and hands-on helps of the measurement system in Chapter 5, his advanced insights and worked out solutions on the modeling and equivalent circuit in Chapter 6, the writing style in all chapters, and helping me to realize and correct the blind spots in my thought; Professor Philip J. Bos, for the guidance in device modeling and circuit simulation; Professor Mukerrem Cakmak and Dr. Tsang-Min Huang, for understanding and implementing stretching machine in their lab at the University of Akron; Mr. Merrill M. Groom, for sharing his expertise spanning from mechanical apparatus, software programming, to electronic circuits, and his help from initial design, debugging, to final implementation.
I would also like to thank the Professors who taught me in their courses: Professor David W. Allender, Professor Liang-Chy Chien, Professor Antal I. Jakli, Professor Chanjoong Kim, Professor Peter Palffy-Muhoray, Professor Jonathan V. Selinger, Professor Robin L. Selinger, and Professor Deng-Ke Yang. From their courses, I have learned the secret of liquid crystal and how to distinguish the truth and the false from the plenty knowledge and information we access every day.

I would also like to thank many friends and fellow students with whom I have shared parts of this journey, in the lab, in the classroom, and out in the real world. To name a few: Shinying, Shawn, Christopher, Nick, Volodymyr, Liwei, Lulu, Rafael, Jason, Jie, Yue, Lei, Hossein, Huan, Cheng, Paul, Junren, Farhadi and my entire incoming class (Mykhailo, Hui, Nik, Emine, Youngki, Shuojia, Cuiyu, and Shuang) for sharing the bitter and sweet two years of tough LCI courses on which our core competences were built.

Finally, I would like to thank my family and my wife Chung-Pin Wang for their love and support for not just the past 5 years, but all my entire life.

Da-Wei Lee

20 March 2015, Kent, Ohio, USA
For my wife Chung-Pin Wang,

who married me, stayed with me, and supported me in the past 15 years.
CHAPTER 1

Introduction

People’s life are changed and improved by new products based on new technology. The current smart phone “revolution” is the latest example. Another example is the consumer digital camera, after the first introduction in mid 1990s [1−2], nowadays most people do not use traditional camera and photographic film anymore [3−4]. This is because digital cameras and flash memory are much more convenient, compact and adjustable compared with old solutions. Another example is the liquid crystal (LC) lens [5−7] where focal length is adjusted by charging an externally applied voltage. LC lens are much more convenient, compact and adjustable compared with traditional glass lens, because the parabolic phase profile can be adjusted electronically, rather than mechanically. The thickness of the lens is therefore shrunk dramatically and the function of the lens is extended significantly. Although LC lens are still in the development stage, but we believe that it will replace traditional lens in the near future. Similarly, in this study, we developed a new technology which can be used in PDLC windows and now recognize many other applications. The transparent areas of the new PDLC window can be selectively controlled by external voltages and frequencies. This new PDLC window functions as an electronic venetian blind.
1.1 Transparent Conductive Material

1.1.1 Importance of Transparent Conductive Material

As the name implies, transparent conductive material (TCM) is optically transparent to allow the visible light to pass, and is also electrically conductive. TCMs are essential for flat panel displays, touch screens, and solar cells. As shown in Figure 1.1 [8], the use of TCM as common electrode and display electrode in Thin Film Transistor Liquid Crystal Display (TFT LCD) is clearly illustrated. If the electrodes in display and touch panel are not transparent, the light of images could not reach the eyes of the user.

![Figure 1.1: Structure and components of TFT LCD [8].](image)

TCMs can be categorized into inorganic and organic [9]. Inorganic TCMs are typically metal oxide compound [10–11], for instance, indium tin oxide (ITO), indium
oxide (In$_2$O$_3$), tin oxide (SnO$_2$) and zinc oxide (ZnO). Organic TCMs are typically conductive polymer formulation such as PEDOT.

### 1.1.2 Why ITO

As shown in Figure 1.2, more than 95% of TCM used in display, touch panel, and solar cell products is ITO nowadays [12–13], which is the reason we choose ITO as the raw material in this study. ITO is the standard by which other TCM candidates will be measured.

![Market share of different transparent conductive materials](image)

**Figure 1.2:** Current and forecast market share of different transparent conductive materials [12].

### 1.1.3 Electrical and Optical Properties of ITO

ITO, indium tin oxide, or tin-doped indium oxide, is a weight combination of ~90% indium (III) oxide (In$_2$O$_3$) and ~10% tin (IV) oxide (SnO$_2$) [14–16]. It is a highly doped n-type semiconductor in which oxygen is replaced by tin dopants, and a low electrical resistivity of 2 ~ 4 × 10$^{-4}$ ohm·cm is achieved, because the Fermi level [15, 17] is located above the conduction level [18]. The carrier concentration of ITO is 10$^{20}$ ~ 10$^{21}$ cm$^3$ [16, 19], which is 2 to 3 orders higher than the degenerate doping concentration,
$>10^{18}$ cm$^3$, for intrinsic silicon which has $5 \times 10^{22}$ atoms/cm$^3$ [20]. ITO is wide band gap ($E_g = 3.5 \sim 4.3$ eV) semiconductor [16, 18], which makes it transparent for the light in the visible and near infrared regions, because the photon energy is much less than the band gap energy, as shown in Table 1.1.

<table>
<thead>
<tr>
<th></th>
<th>Infrared</th>
<th>Visible</th>
<th>Ultraviolet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>100 ~ 1 μm</td>
<td>0.7 ~ 0.4 μm</td>
<td>0.1 ~ 0.01 μm</td>
</tr>
<tr>
<td>Frequency (Hz)</td>
<td>3×10$^{12}$ ~ 3×10$^{14}$</td>
<td>4×10$^{14}$ ~ 7×10$^{14}$</td>
<td>3×10$^{15}$ ~ 3×10$^{16}$</td>
</tr>
<tr>
<td>Photon Energy (eV)</td>
<td>0.012 ~ 1.2</td>
<td>1.7 ~ 3</td>
<td>12 ~ 120</td>
</tr>
</tbody>
</table>

Table 1.1: Photon energy calculated in near infrared, visible, and ultraviolet regions.

**Frequency** = light speed ($3\times10^8$ m/sec) / wavelength. **Photon energy** = Planck constant $h$ ($4\times10^{-15}$ eV · sec) × frequency.

Having a clear understanding of the physical meaning in these numbers of electrical and optical properties of ITO is very useful. For instance, the sheet resistance [21] of commercially available ITO on PET film is always provided by the supplier, therefore we can calculate the thickness of ITO layer by using the value of electrical resistivity of $2 \sim 4 \times 10^{-4}$ ohm·cm mentioned above. An example of this calculation is shown as follows. For an ITO on PET film, the sheet resistance is 23 ohm/square and the electrical resistivity is $4.58 \times 10^{-4}$ ohm·cm [16]. Based on the definitions of sheet resistance and electrical resistivity, the thickness of ITO layer should be:

$$
\left( \frac{1cm}{ \text{thickness} } \right) \cdot (4.58 \times 10^{-4} \Omega) = 23 \Omega \Rightarrow \text{thickness} = 10^{-2} m \cdot \frac{4.58 \times 10^{-4}}{23} \approx 2 \cdot 10^{-1} \mu m = 200 \mu m
$$

This value is exactly the thickness of ITO layer [16].
The optimized electrical and optical properties of ITO could be achieved by adjusting the weight percentage of tin oxide, as shown in Figure 1.3 [10], in which the sheet resistance, 550nm green light transmission, and electron mobility are all optimized at 20% tin oxide.

![Chart showing electrical and optical properties of ITO](image)

**Figure 1.3**: Optimization on both the electrical and optical properties of ITO by adjusting the weight percentage of tin oxide [10].

The transmission spectrum of ITO is shown in Figure 1.4 [16], more than 90% of incident light passes through 200 nm thick ITO film in the region of 400 nm ~ 1300 nm, which covers the visible light region of 400 nm ~ 700 nm. One interesting thing is that the transmission decreases at wavelength longer than 1400 nm, and it is strongly determined by the annealing temperature; this behavior may be a solution for “transparent and reducing solar heat” windows used in vehicles or buildings.
Figure 1.4: Transmission and reflection spectrum of thickness 200 nm ITO films treated by different anealing temperatures [16].

1.2 Glass Substrate versus Plastic Substrate

The thickness of ITO layer is around 200 nm for many commercial applications, it is too thin and therefore a thicker substrate is required to hold the ITO layer. There are two choices for the substrate of ITO, glass and plastic film. Glass substrates are the conventional solution, but it is gradually being replaced by plastic films which have much less weight, elasticity, and bending ability for new products in new applications.

In the past 10 years, there were many research activities focused on developing new displays made by plastic films, instead of glass substrates [22–23]. The first demonstration of an OTFT active-matrix liquid-crystal display, and also the first demonstration of a TFT active-matrix liquid-crystal display of any type fabricated on a polyester substrate were reported by Professor John L. West et al. in 2001 [24]. After
that, the prototype of OTFT-driven OLED flexible display which could reproduce dynamic images while being repeatedly wrapped around a cylinder with 4 mm radius more than 1000 times was demonstrated by Sony Corporation on May 2010, as shown in Figure 1.5 [25].

Figure 1.5: In May 2010, Sony Corporation demonstrated 80 µm thickness 4.1 inch wide full color OTFT-driven OLED flexible display made by 20 µm flexible substrates, which can be wrapped around by a cylinder with 4 mm radius [24].

In order to construct high performance TFT on plastic film, the bond–debond process [26] was developed to solve the dimension variation issue of plastic film during conventional TFT manufacturing processes around 200°C. Plastic film temporarily bonded with glass substrate is handled together to go through the TFT processes, and after that, plastic film is de-bonded from glass substrate.

Unfortunately, there is no commercialized product of plastic film display available in the market yet; one major problem is the water and oxygen isolation issue of plastic film [27–28], which limits the life time of OLED.
Although glass substrate is not practically replaced by plastic film in display application, but plastic film had been proved to be a much better solution comparing with glass substrate in other products, for instance, polymer dispersed liquid crystal (PDLC) [29] window, LCD writing tablet Boogie Board™ [30], resistance touch panel [31], and the cracked ITO film studied in this paper.

PDLC was invented in Liquid Crystal Institute, Kent State University and then commercialized in privacy window application by several companies around 1985 [32]. The principle of PDLC window is shown in Figure 1.6. Several microns liquid crystal droplets are surrounded by polymer, which is sandwiched by ITO electrodes in between around 20 microns gap is defined by spacers.

In the off state, no electric filed is applied between ITO electrodes, the orientation (i.e. director) of liquid crystal molecules inside each droplet are different in different droplets. When the incident light of electromagnetic wave having a specific polarization [33] perpendicularly enters into PDLC, the refractive index [34] inside each droplet (with respect to the incident light) is determined by the director. Because the directors in the droplets are randomly distributed, so most of the refractive index inside the droplets is different from the refractive index of polymer surrounding the droplets. From Snell Law, we know that the incident light will be refracted at the interface between liquid crystal and polymer. There are thousands of micron size droplets distributed in (10 micron)$^3$ volume of PDLC, so thousands time of refractions occur when the incident light is
passing through PDLC, the light is scattered, and an opaque milky white state is observed in the other side.

In the on state, electric field is applied between ITO electrodes, the liquid crystal molecules are uniformly parallel-aligned with electric field inside each droplet. When the incident light of electromagnetic wave having a specific polarization perpendicularly enters into PDLC, the refractive index inside each droplet (with respect to the incident light) is determined by the director. Because the directors in all droplets are uniformly parallel-aligned, so the refractive indexes inside the droplets are identical to n₀. The refraction index of polymer surrounding the droplets is designed to be n₀. The refractive index inside all droplets and the refractive index of polymer surrounding the droplets are identical. From Snell Law, we know that the incident light will not be refracted and just pass through at the interface between liquid crystal and polymer, and a transparent state is observed along the perpendicular direction in the other side.

![Figure 1.6: The working principle of PDLC [35].](image)

PDLC window was made between glass substrates in the early years, but it became more popular after replacing the glass substrate into plastic film, which made it
more convenient, compact, and adjustable. As shown in Table 1.2, the advantages of plastic film for PDLC window products are clear, and therefore new applications like laminating plastic PDLC film on the windows of vehicles are thriving in the market nowadays.
<table>
<thead>
<tr>
<th>PDLC Window</th>
<th>Glass Substrate</th>
<th>Plastic Film</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness</td>
<td>1–2 mm</td>
<td>100–200 μm</td>
</tr>
<tr>
<td>Density</td>
<td>2.65 g cm⁻³</td>
<td>1.38 g cm⁻³</td>
</tr>
<tr>
<td>Weight Ratio for Identical Size</td>
<td>20</td>
<td>1</td>
</tr>
</tbody>
</table>

| Building Application | * High cost: replace old windows.  
|                     | * Inconvenient: hard to handle, large and heavy plates.  
|                     | * Low cost: laminate on old windows.  
|                     | * Convenient: easy to handle, compact and light rolls.  |

| Vehicle Application | Not adjustable to be laminated on curved window surface.  
|                     | Adjustable to be laminated on curved window surface.  |

Table 1.2: Comparing glass substrate with plastic film in the PDLC window applications.

Another example is the writing tablet of Cholesteric Liquid Crystal (ChLC). The bistability of planar texture and focal conic texture of ChLC was first discovered in the 1970s [36, 37], but its potential in bistable display application was not fully recognized then. ChLC was first used in bistable reflective display and the bistability was rediscovered and its importance was recognized in the 1990s [38–42]. After that, the research of bistable ChLC display related application has been thriving and its performance has been improved significantly.

The basic principle of ChLC is that the light passing through ChLC can be controlled by the applied electric field, and a more important function is: after a specific electric field is applied on ChLC for several seconds and then turned off, one of two different optical states can be achieved and stayed without applying electric field, which means there are two stable states, i.e. “bistable”, existed in ChLC.
As shown in Figure 1.7 (a), planar texture and focal conic texture of ChLC are illustrated. In the planar texture, circularly polarized light is reflected, but in the focal conic texture, light is scattered in forward directions. The planar texture can be switched into the focal conic texture by applying a low-voltage pulse, and the focal conic texture can be switched into the planar texture by applying a high-voltage pulse, as shown in Figure 1.7 (b).

Figure 1.7: Yang [43], (a) Cholesteric Liquid Crystal (ChLC) device structure and two stable optical states while no electric field is applied between two layers of ITO. (b) The bistable behavior of ChLC: reflection versus applied voltage V, the reflection is measured after applying V between two layers of ITO for several seconds and then turned off. There is no voltage applied on CHLC layer while the reflection is measuring.
The bistability of Cholesteric Liquid Crystal (ChLC) and its application in reflective display was studied since 1990 [43]. In 1995, the writable function of Cholesteric Liquid Crystal Display (ChLCD) was first reported by Professor John L. West [44]. Since 2006, the writable function of ChLCD was studied in Industry Technology Research Institute (ITRI), Taiwan [45–47]. From 2007 to 2010, Kent Displays Inc. (KDI) did research on the writing tablet made by ChLCD [48]. In 2010, KDI started to sell a new product named Boogie Board™, a ChLCD writing tablet on which a plastic covering film is required, because the pressure released from the tip of user’s stylus has to be precisely transferred into the cholesteric liquid crystal layer underneath, as shown in Figure 1.7; and therefore the elastic plastic film is much better than the rigid glass substrate in this application, because the pressure can’t be sensitively transferred to the liquid crystal layer through a rigid surface. By the end of 2012, more than 1 million units of Boogie Boards™ had been sold to the customers, so the revenue is more than 30 million dollars based on the unit price $30. This is clear evidence that plastic film is much better than glass substrate in some new applications.

Another benefit of PDLC or ChLC display is that it does not need polarized light to be operated, so birefringence plastic PET film can be used as the substrates, which makes the display much more light and flexible.
Figure 1.8: The working principle of Boogie Board\textsuperscript{TM}, on which the finger tip or stylus releases pressure on the surface and deform the plastic film which is elastic, therefore the cholesteric liquid crystal underneath is changed into planar state [48].

1.3 Cracked ITO

The cracking of brittle thin films on a semi-flexible substrate has been investigated many times and produced several theories to model the formation and propagation of these cracks [49–50]. For example early studies investigated the formation of cracks in glazes on ceramic pottery. More recently studies include the investigation of thin films of ITO coated on flexible substrates and in particular ITO coated on PET. This is because ITO is by far the most common material used as the transparent, conducting electrodes used in flexible displays and solar cells. PET remains the most robust flexible substrate for many electro-optic applications.

In all of these studies the cracking of the ITO has been viewed as a limitation and problem need to be avoided. This thesis investigates how the cracking of the ITO can be used to form useful electrodes for a variety of applications and how the cracking can be controlled and enhanced to produce optimum electrical properties for a variety of applications, including switchable windows and touch screens.
Based on the study in section 1.1 and 1.2, we can realize why ITO coated on plastic film is an important commercialized solution for many new products nowadays, and it provides unique values required in each specific application. But still, ITO plastic film is not perfect, because ITO is fragile and brittle, which is a major problem frustrating the commercialization of flexible displays.

Many studies had been done to analyze the brittle and fragile property of ITO coated on plastic film. For example, multiple cracking of platinum deposited on a PET substrate under tensile deformation was studied by Volynskii, et al [51]. The thin platinum film fractured upon stretching of the substrate forming long cracks perpendicular to the elongation direction of the PET substrate. The separation of the cracks was on the order of 5~10 microns and depended on the thickness of the platinum and tensile stress applied on the PET substrate.

A mandrel-bending test system was developed to evaluate the degradation of ITO/PET by Crawford, et al [52]. They found that even when the strain was below the virgin cracking threshold, there were measurable changes in ITO resistance, and cyclic loading of ITO/PET showed three regimes of resistance increase. Koniger, et al [53] observed an oscillating change in resistance for bending radii smaller than 14 mm which could be explained by an alternating extension of cracks under the applied oscillatory tensile stress. They also found that the cracks extended across the width of the substrate forming perpendicular to the stress.
In the above mentioned studies, the cracking of the ITO was viewed as a disadvantage to be avoided. In this thesis, we have found on controlling and optimizing the cracking process to produce unique and useful substrates. In this study, we explored how the formation of uniform cracks in the ITO coated on a PET substrate could be exploited to produce uniform line electrodes. Inspired by the bending techniques reported by Crawford [52] and Koniger [53], we constructed a simple apparatus to uniaxially bend ITO coated PET substrates. We used the resulting substrates to make a PDLC shutter where individual lines could be switched without resorting to photolithography or printing.

1.4 Heat Stabilized PET

ITO coated on PET substrate is the most common commercial available product of transparent conductive material coated on plastic substrate nowadays, and they are heat stabilized before selling to the end customers. In section 2.3, our cracked ITO on PET film is performed at temperature 100°C with stretching loading 60~80 kg in a highly instrumented stretching system, as shown in Figure 2.13. So here we need to study the general background of heat stabilized PET, and therefore to understand the effect caused by the heat stabilized PET on the behavior of heated and stretched cracked ITO PET film in our study.

Heat stabilized PET means it becomes more insensitive (i.e. less strain response) under the same stimulation of heat or stress. PET must be stretched within the rubbery region to an extent which results in strain hardening [54]. Stretching PET in the range
between two temperatures, i.e. glass transition temperature \(T_g\), around 80 °C) and crystallization starting temperature (around 130 °C), results in the molecules to be aligned to an orientation, which leads to strain-induced crystallization. The tensile modulus of oriented PET film is 5 times larger than that of non-oriented PET film [55].

One commercialized process of biaxial stretched PET is to stretch it along the machine direction first, and then stretched it along the transverse direction [56], the resulting film can be heated to get the strain hardening property. Another way of getting the strain hardening property is to use the simultaneous biaxial stretching process [57].

1.5 Goal and Outline

In this work, we will present the result of a novel smart window technology, which establishes a revolutionary, simple, and low cost manufacturing method to make parallel cracked ITO stripes on flexible PET substrate. It successfully transfers the drawback of fragile ITO into a commercial manufacture technology. Flexible PET substrate coated by thin layer of ITO is processed to create parallel ITO cracks, and the resulting cracked ITO on PET is used as a substrate in the polymer dispersed liquid crystal (PDLC) window. It creates a virtual “venetian blind” effect, where the amount of light admitted passing through the window is controlled by frequency and amplitude of applied voltage. This work has produced 3 US patent applications and the launch of a start-up company, Flexible ITO Solutions (FITOS) Inc., in 2013.

CHAPTER 2 describes the origin of utilizing cracked ITO on PET substrate and all experiments attempted to enhance the cracks, including the stretching process
conducted in Akron University. Following that, CHAPTER 3 points out the issues of stretching process, and provides an innovative approach of solving the issues of stretching process by a cost effective and reproducible manufacturing process. Several real size PDLC windows made by Cracked ITO on PET substrate are successfully implemented, and prove that our technology is real, low cost, and reproducible. CHAPTER 4 discusses about the basic physics of cracked ITO on PET substrate, in which the previous studies of cracks are reviewed thoroughly, and used to calculate the situation in our case. CHAPTER 5 is a unique and beautiful discovery found in this study: the frequency dependent line with of driving image. The frequency dependent driving image is observed and measured in PDLC samples, and the sample size is also dependent on the line width of driving image. This discovery provides a new approach of driving displays which may lead to new applications. CHAPTER 6 provides the theoretical explanation on the frequency dependent line width of driving image. A circuit model of the PDLC device made by cracked ITO on PET substrate is established, therefore analytical calculation and computer simulation are both provided to explain the frequency dependent behavior, and now we know that it is basically a low pass filter. Finally, our discoveries are summarized in CHAPTER 7.
CHAPTER 2

Parallel Cracked ITO Made by Stretching Process

2.1 Initial Idea of Cracking ITO

The birth of cracking ITO idea in this dissertation was from an unexpected discovery of reviewing SEM pictures. In summer 2011, Professor John L. West assigned me a research project of studying different PDLC morphologies caused by different surfaces, for instance, PET surface and PEDOT:PSS i.e. poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) surface. I reported him several SEM pictures of “PEDOT spin-coated on ITO on PET substrate” and “PEDOT spin-coated on PET substrate”, as shown in Figure 2.1. The cutting edge and surface of PEDOT were observed by using SEM.
Figure 2.1: Long-lasting parallel cracks appeared near the cutting edge of PEDOT on ITO or PET substrate. (a) (c) (e) are SEM pictures of “PEDOT on ITO on PET” sample with magnification of 500, 1000, and 2500 respectively. (b) (d) (f) are SEM pictures of “PEDOT on PET” sample with magnification of 500, 1000, and 2500 respectively.

Here we find that a problem may become to a solution. As shown in Figure 2.1 (b) (d) (f), for the “PEDOT on PET” sample, there is no crack found on PEDOT around the cutting edge; for the “PETDOT on ITO on PET” sample, Figure 2.1 (a) (c) (e), two kinds of cracks on PEDOT are observed: one is parallel and the other one is perpendicular with the long direction of cutting edge. A key feature of the perpendicular
PEDOT cracks is that they are all long lasting and parallel with each other without any intersection. Professor West told me this is interesting and we should study and try to utilize it. Because these long lasting and parallel PEDOT cracks should be caused by the cracking of ITO underneath. If it is true, there should be also long-lasting and parallel ITO cracks produced in ITO layer, which may be useful as a new manufacturing method of producing parallel ITO stripes without photolithography and wet-etching processes, it means a problem of crack may become to a solution of electrode patterning.

As shown in section 1.3, there were many studies had been done to analyze the behavior of cracked ITO on flexible substrate, all of these studies treated cracked ITO as a drawback which should be avoided. No one has ever thought about the possibility of utilizing crack ITO, which is what we want to do. We want to transfer the drawback of fragile ITO into a new solution which provides simple, low cost, and reproducible manufacturing method of making parallel cracked ITO stripe electrodes useful in display products like PDLC window, passive matrix display, and touch panel.

We will try four methods to make parallel cracks on PET substrates, as summarized in Table 2.5, and then the stretching process will be discussed in section 2.3.

The first method of making parallel cracks on PET substrate is shown in Figure 2.2 (a), a sample of 2 cm × 8 cm ITO on PET substrate is prepared. It is manually attached on the surface of a rectangle corner of a table, and then smoothly moved along two orthogonal directions as shown in Figure 2.2 (b). The solid rectangle corner of the table and the pulling forces from hands provide a stress applying on ITO on PET
substrate, and it produces a series of ITO cracks as shown in Figure 2.2 (c) to (f), which are observed with different magnifications under an optical microscope.
Figure 2.2: The first try of making parallel ITO cracks on PET substrate. A piece of 2 cm × 8 cm ITO on PET film (a) is manually attached and moved at a rectangle corner of a table (b) with ITO side on top. The resulting cracks in ITO are observed under optical microscope, as shown in (c) to (f).

As shown in Figure 2.2 (c) to (f), two features of these cracks in ITO can be observed. First, 70% cracks are long lasting and parallel with neighboring cracks without having interactions, and 30% cracks are not parallel with neighboring cracks, which means there are intersections occurred. Second, for high magnification pictures (e) and (f), there are slight ITO cracks can be observed between obvious ITO cracks, where the separation between slight ITO cracks is around 10 microns, and the separation between obvious ITO cracks is around 30 to 40 microns, which means around 2 to 3 slight ITO cracks are distributed between two neighboring obvious ITO cracks.

The resistance measurement of the first tried ITO cracks was also conducted. Because these ITO cracks are basically parallel with each other, so the resistance perpendicular to ITO cracks must be much higher than the parallel one. As shown in Table 2.1, the perpendicular resistance increases 45 times which is much larger than the 5 times increasing of parallel resistance.

<table>
<thead>
<tr>
<th></th>
<th>( R_{AB} ) (ohm)</th>
<th>( R_{CD} ) (ohm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Before Cracking</td>
<td>260 ~ 270</td>
<td>110 ~ 120</td>
</tr>
<tr>
<td>After Cracking</td>
<td>12,000</td>
<td>600 ~ 700</td>
</tr>
<tr>
<td>Resistance Increase</td>
<td>45 times</td>
<td>5 times</td>
</tr>
</tbody>
</table>

Table 2.1: The perpendicular resistance \( R_{AB} \) and the parallel resistance \( R_{CD} \) are measured by using multi-meter on ITO on PET film as shown in Figure 2.2 (a).
2.2 Issue of Cracking ITO

Based on two features observed in Figure 2.2 (c) to (f), there are issues of the first tried ITO cracks: How to avoid the intersection between two neighboring ITO cracks? This will increase the parallel resistance which we want it to be as small as possible. How to make the parallel cracks wider and significant? This will increase the perpendicular resistance which we want it to be as large as possible. Ideally, we want to increase the perpendicular resistance to be infinite, which means two neighboring cracked ITO stripes are completely and electrically isolated; and the parallel resistance is maintained at the original conductivity of ITO, which means there is no intersection between two neighboring cracks and all cracks are parallel with each other. In other words, we want the value of ratio $R_{AB} / R_{CD}$ to be as large as possible.

So we try out new method of making parallel ITO cracks on PET substrate: use a tight radius, about 2 mm, of bending curvature. As shown in Figure 2.3, two pieces of 5 mm thickness glass plate are separated by sphere beads of 4 mm diameter $D$. A sample of 4 cm $\times$ 8 cm ITO on PET substate film is bent between two glass plates, and then the top plate is manually moved along the direction perpendicular to the bending edge of the sample with a speed around 1 cm / sec, while the bottom plate is fixed at the same place. The thickness of ITO on PET substrate is 150 microns, so the bending radius $R$ is equal to $\frac{D - 2 \cdot 150 \mu m}{2}$ which is 1.85 mm, about 2 mm.
Figure 2.3: The second try of making parallel ITO cracks on PET substrate. (a) A manually bending apparatus designed to make parallel cracked ITO stripes on PET substrate. Sphere beads of diameter 4mm are distributed between two glass plates to maintain a gap of 4 mm. Top plate is manually moved with a speed of 1 cm / sec, while the bottom plate is fixed. (b) The real apparatus designed in (a). (c) The resulting cracked ITO on PET substrate, which is pulled to be straight. (d) The resulting cracked ITO on PET substrate, which is free standing as a roll of plastic film.

The sample of ITO on PET substrate is bent and rolled 6 times, in which one time rolling process means rolling forward and rolling backward. The resistance measurement result is shown in Table 2.2, in which a new way of measuring resistance on a bent ITO surface on PET substrate is also introduced. A new technology is discovered [58] by analyzing these values of measured resistance: a piece of cracked ITO on PET substrate can be used as a device which can measure the bending curvature of the tested surface by monitoring the value of resistance perpendicular to cracked ITO stripes. After cracking,
the value of $R_{AB}$ measured on flat surface is 8k ohm, and this value is increased 37.5 times to 300k ohm when it is measured on curved surface of bending radius 1.5 cm.

![Diagram showing crack and resistance measurement](image)

<table>
<thead>
<tr>
<th>Resistance measured on flat surface</th>
<th>Resistance measured on curved surface</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_{AB}$ (ohm)</td>
<td>$R_{CD}$ (ohm)</td>
</tr>
<tr>
<td>Before Cracking</td>
<td>~100</td>
</tr>
<tr>
<td>After Cracking</td>
<td>~8k</td>
</tr>
<tr>
<td>Resistance Increase</td>
<td>80 times</td>
</tr>
<tr>
<td>$R_{AB}$ (ohm)</td>
<td>~100</td>
</tr>
<tr>
<td>$R_{CD}$ (ohm)</td>
<td>~0.8k</td>
</tr>
<tr>
<td>Resistance Increase</td>
<td>8 times</td>
</tr>
<tr>
<td>$R_{AB}$ (ohm)</td>
<td>~100</td>
</tr>
<tr>
<td>$R_{CD}$ (ohm)</td>
<td>~300k</td>
</tr>
<tr>
<td>Resistance Increase</td>
<td>3k times</td>
</tr>
<tr>
<td>$R_{AB}$ (ohm)</td>
<td>~100</td>
</tr>
<tr>
<td>$R_{CD}$ (ohm)</td>
<td>~20k</td>
</tr>
<tr>
<td>Resistance Increase</td>
<td>200 times</td>
</tr>
</tbody>
</table>

Table 2.2: The measurement of resistance $R_{AB}$ and $R_{CD}$ on flat surface and curved surface, before and after cracking ITO on PET substrate by using a manually bending apparatus with tight radius 2 mm of bending curvature.

The effectiveness of tight radius 2 mm of bending curvature as shown in Figure 2.3 is better than the first try of orthogonal forces as shown in Figure 2.2, because the
perpendicular resistance $R_{AB}$ is increased 80 times and 45 times, respectively; and the parallel resistance $R_{CD}$ is increased 8 times and 5 times respectively; so the ratio $\frac{R_{AB}}{R_{CD}}$ is improved from $\frac{(12000/4)}{650} = 4.62$ to $\frac{8k}{0.8k} = 10$, which is roughly 2 times improvement.

As shown in Figure 2.2 (e), the length AB is 4 times longer than the length CD, so $R_{AB}$ needs to be divided by 4 in this calculation.

2 times improvement is good. But we want it to be much better. So we keep on trying out new method of making parallel ITO cracks on PET substrate: cracked ITO is etched by 2.5 wt % HCl in H$_2$O. The reason of why chemical etching might work is that the ITO may be etched completely along the vertical direction and meanwhile there are still ITO remained along the horizontal direction. As shown in Figure 2.4, after the sample of ITO on PET substrate is placed around the surface of a glass cylinder with 3 cm diameter as shown in Figure 2.4 (a). After 6 times of rolling, the sample “holds” the glass cylinder by itself, and doesn’t need any other helps. Then it is placed into the solution of 2.5 wt % HCl in H$_2$O for a period of time, and measure $R_{AB}$ and $R_{CD}$ at the end of each period of time, as shown in Table 2.3. We intend to etch the cracked ITO while it is bent as a curved surface, because the measured $R_{AB}$ in Table 2.2 suggests that the isolation between neighboring ITO stripes is increased when it is bent as a curved surface from a flat surface.
Figure 2.4: The third try of making parallel ITO cracks on PET substrate. (a) The cracked ITO on PET attached on the curved surface of 3 cm diameter cylinder. (b) The curved cracked ITO on PET substrate is immersed in the 2.5 wt % HCl in H2O solution.

<table>
<thead>
<tr>
<th>Total Etching Time (minutes)</th>
<th>R_{AB} (k ohm)</th>
<th>R_{CD} (k ohm)</th>
<th>R_{AB} (k ohm)</th>
<th>R_{CD} (k ohm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>8</td>
<td>0.8</td>
<td>300</td>
<td>20</td>
</tr>
<tr>
<td>2</td>
<td>15</td>
<td>1</td>
<td>300</td>
<td>20</td>
</tr>
<tr>
<td>4</td>
<td>15</td>
<td>1</td>
<td>300</td>
<td>20</td>
</tr>
<tr>
<td>4 (Rest 24 hours)</td>
<td>50</td>
<td>1</td>
<td>700~1.4k</td>
<td>20~90</td>
</tr>
<tr>
<td>6</td>
<td>50</td>
<td>1</td>
<td>700~1.4k</td>
<td>20~90</td>
</tr>
<tr>
<td>8</td>
<td>80~100</td>
<td>1~3</td>
<td>&gt;2k</td>
<td>200~800</td>
</tr>
<tr>
<td>10</td>
<td>80~100</td>
<td>1~3</td>
<td>&gt;2k</td>
<td>200~800</td>
</tr>
<tr>
<td>12</td>
<td>150~300</td>
<td>5~10</td>
<td>&gt;2k</td>
<td>1~2k</td>
</tr>
<tr>
<td></td>
<td>12 (Rest 24 hours)</td>
<td>300</td>
<td>7-10</td>
<td>&gt;2k</td>
</tr>
<tr>
<td>------</td>
<td>--------------------</td>
<td>-----</td>
<td>------</td>
<td>-----</td>
</tr>
<tr>
<td>14</td>
<td>1-2k</td>
<td>10-30</td>
<td>&gt;2k</td>
<td>&gt;2k</td>
</tr>
<tr>
<td>16</td>
<td>&gt;2k</td>
<td>15-40</td>
<td>&gt;2k</td>
<td>&gt;2k</td>
</tr>
</tbody>
</table>

Table 2.3: The measurement of resistance $R_{AB}$ and $R_{CD}$ after the process of “Wet etching of cracked ITO on PET substrate” in which PET substrate is bent as a curved sample.

Based on the measured resistances in Table 2.3 and Table 2.2, we find that the “bending and wet etching” method is better than “bending only” method, because the ratio $\frac{R_{AB}}{R_{CD}}$ is improved from $\frac{8k}{0.8k} = 10$ (as shown in Table 2.2) to $\frac{(1000k}{10k}) \div \frac{(50k}{15k}) = 30 \sim \frac{(2000k}{30k}) \div \frac{(50k}{15k}) = 20$ (as shown in Table 2.3), which is 3~2 times improvement, the explanation is as follows.

The maximum value of $\frac{R_{AB}}{R_{CD}}$ occurs after etching 14 minutes, while $R_{AB}$ is measured as 1000k to 2000k ohm and $R_{CD}$ is measured as 10k to 30k ohm. The reason why it is required to be divided by $\frac{50k}{15k}$ is because this factor of improvement is contributed by the process of resting 24 hours, rather than etching 14 minutes. After etching 4 minutes and 12 minutes, a process of resting 24 hours is conducted respectively. One thing very interesting is observed: after the first 24 hours resting process, $R_{AB}$ is increased from 15k ohm to 50k ohm (i.e. 3.33 times), but $R_{CD}$ stays at
around 1k ohm. It means the PET substrate is extended along AB direction after 24 hours, and this behavior might be related to the heat stabilized process and molecule structure discussed in page 33 to 36.

The second maximum value of $\frac{R_{AB}}{R_{CD}}$ occurs after etching 8 minutes, while $R_{AB}$ is measured as 80k to 100k ohm and $R_{CD}$ is measured as 1k to 3k ohm, so the ratio $\frac{R_{AB}}{R_{CD}}$ is improved from $\frac{8k}{0.8k} = 10$ to $(\frac{80k}{1k}) \div (\frac{5k}{15k}) = 24 \sim (\frac{100k}{3k}) \div (\frac{5k}{15k}) = 10$, which is roughly 2.4 ~ 1 times improvement.

3.33 times improvement of resting 24 hours, or 3~2 times improvement of 14 minutes etching by HCl, or 2.4~1 times improvement of 8 minutes etching by HCl, all of them are good trials. Although chemical wet etching can improve the resistance ratio as we expected, but we want to find a better method, because from practical point of view, the chemical etching will make the process more complicated and increase the manufacturing cost. So we keep on trying out new method of making parallel ITO cracks on PET substrate: cracked ITO is heated and loaded by a loading in a heat oven. As shown in Figure 2.5, after the sample of ITO on PET substrate is bent and rolled 6 times, it is placed in a heat oven, which is heated from 25 to 160 °C.

In order to implement the idea of “heating and loading” process, a practical solution need to be figured out. As shown in Figure 2.5 (a), two pieces of glass substrate are adhered with both edges of cracked ITO sample by using 5 minutes epoxy, four clips
are used to enhance the adhesion between glass substrates and the sample. As shown in Figure 2.5 (b), the sample is placed in the central of a heat oven, in which the temperature can be controlled. Two multi-meters are used to measure the resistance of $R_{AB}$ and $R_{CD}$. As shown in Figure 2.5 (c), two clips on top prevent the whole system falling down, and the bottom glass substrate and the 429 grams metal block are attached by using two rubber bands.
Figure 2.5: The fourth try of making parallel ITO cracks on PET substrate. (a) Two pieces of glass substrate and cracked ITO sample are adhered by using 5 minutes epoxy and four clips. (b) Two multi-meters are used to measure the resistance of $R_{AB}$ and $R_{CD}$, while cracked ITO sample is heated. (c) Cracked ITO sample is heated while loading 429 grams metal block.

As shown in Table 2.4, the temperature in the heat oven is increased from 25°C to 160°C, and stayed at 100°C to 160°C for 3 to 5 minutes. Based on the measured resistances, we find that the “heating and loading” method can improve the ratio $\frac{R_{AB}}{R_{CD}}$

from $\frac{3.83k}{0.4k} = 9.58$ to $(\frac{13.49k}{0.35k} = 38.54 \sim \frac{16.88k}{0.35k} = 48.23)$, which is roughly 3~4 times improvement. The maximum value of $\frac{R_{AB}}{R_{CD}}$ occurs at heating temperature of 111~114°C, while $R_{AB}$ is measured as 13.49k to 16.88k ohm and $R_{CD}$ is measured as 0.35k ohm.
The temperature dependent behavior of $\frac{R_{AB}}{R_{CD}}$ is as follows. In the range from 25°C to 110°C, there is no big change. In the range from 111°C to 117°C, $\frac{R_{AB}}{R_{CD}}$ stay around 30 to 40. When the temperature is increased to 120°C, suddenly $\frac{R_{AB}}{R_{CD}}$ is shrunk back to 10, which is very interesting. In the range from 120°C to 160°C, there is no significant change. After 160°C, the rubber band is broken. We tried the “Heating and Loading” process for several times, the results are not identical each time, but basically their behavior follows the same pattern described above.
<table>
<thead>
<tr>
<th>Time (min)</th>
<th>Temperature (C)</th>
<th>$R_{AB}$ (k ohm)</th>
<th>$R_{CD}$ (k ohm)</th>
<th>$R_{AB}/R_{CD}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>25</td>
<td>3.83</td>
<td>0.4</td>
<td>9.58</td>
</tr>
<tr>
<td>10</td>
<td>80</td>
<td>3.84</td>
<td>0.4</td>
<td>9.60</td>
</tr>
<tr>
<td>13</td>
<td>100</td>
<td>4.08</td>
<td>0.41</td>
<td>9.95</td>
</tr>
<tr>
<td>18</td>
<td>100</td>
<td>5.05</td>
<td>0.45</td>
<td>11.22</td>
</tr>
<tr>
<td>23</td>
<td>110</td>
<td>4.58</td>
<td>0.46</td>
<td>9.96</td>
</tr>
<tr>
<td>24</td>
<td>111</td>
<td>11.86</td>
<td>0.35</td>
<td>33.89</td>
</tr>
<tr>
<td>26</td>
<td>114</td>
<td>13.49</td>
<td>0.35</td>
<td>38.54</td>
</tr>
<tr>
<td>27</td>
<td>114</td>
<td>16.88</td>
<td>0.35</td>
<td>48.23</td>
</tr>
<tr>
<td>30</td>
<td>117</td>
<td>10.7</td>
<td>0.35</td>
<td>30.57</td>
</tr>
<tr>
<td>31</td>
<td>117</td>
<td>15.4</td>
<td>0.35</td>
<td>44.00</td>
</tr>
<tr>
<td>32</td>
<td>120</td>
<td>3.6</td>
<td>0.35</td>
<td>10.29</td>
</tr>
<tr>
<td>33</td>
<td>125</td>
<td>3.5</td>
<td>0.35</td>
<td>10.00</td>
</tr>
<tr>
<td>36</td>
<td>130</td>
<td>2.9</td>
<td>0.25</td>
<td>11.60</td>
</tr>
<tr>
<td>38</td>
<td>135</td>
<td>2.6</td>
<td>0.26</td>
<td>10.00</td>
</tr>
<tr>
<td>40</td>
<td>140</td>
<td>2.4</td>
<td>0.26</td>
<td>9.23</td>
</tr>
<tr>
<td>52</td>
<td>138</td>
<td>2.2</td>
<td>0.26</td>
<td>8.46</td>
</tr>
<tr>
<td>61</td>
<td>149</td>
<td>2.2</td>
<td>0.26</td>
<td>8.46</td>
</tr>
<tr>
<td>68</td>
<td>160</td>
<td>2.2</td>
<td>0.26</td>
<td>8.46</td>
</tr>
</tbody>
</table>

Table 2.4: The measurement of resistance $R_{AB}$ and $R_{CD}$ in the process of “Heating and Loading of cracked ITO on PET substrate”.

Why there is a significant (3 to 4 times) increase of $R_{AB}/R_{CD}$ in the temperature range from 111 to 117 degrees, and then it sharply return back to the original level after 120 degree? The reason is related to the heat stabilization, the glass transition temperature, and the crystallization temperature of PET substrate, i.e. the morphology of PET and its thermal property. The morphology-property relationship in oriented PET film was studied by Gohil [110] in 1994, and we can use it to explain what we observed in Table 2.4.

The PET substrate we use here are commercial available product, which is heat-stabilized before selling to us. Comparing with plain PET, the heat-stabilized PET is more resistant to heat, and provides improved adhesive strength and gas barrier ability for
the layer coated on it [59]. That means the PET film is biaxially stretched, and then heat stabilized in a tenter with low tension applied, so the molecule relaxation is allowed and the tendency of PET film to shrink at high temperature is reduced, e.g. shrinkage < 0.1% in air at 150 °C 30 minutes [60]. Heat stabilized PET means it becomes more insensitive (i.e. less strain response) under the same stimulation of heat or stress. PET must be stretched within the rubbery region to an extent which results in strain hardening [54]. Stretching PET in the range between two temperatures, i.e. glass transition temperature (T_g, around 80 °C) and crystallization starting temperature (around 130 °C), results in the molecules to be aligned to an orientation, which leads to strain-induced crystallization. The tensile modulus of oriented PET film is 5 times larger than that of non-oriented PET film [55].

The changes in molecular orientation and percent crystallinity as a function of temperature were studied by Gohil [110]. As shown in Figure 2.6 (a), there are three kinds of molecular orientation in PET film: crystallites (highest order), noncrystalline (medium order), and disordered (lowest order). As shown in Figure 2.6 (b), there are three regimes in which the molecular orientations and percent crystallinity are determined by the heat-set temperature (HST) or annealing temperature; and T_max is the temperature at which the maximum crystallization rate happens, T_m is the melting temperature, 1/t is the rate of relaxation of molecules, G is the rate of crystallization. In regime I, PET film is stretched and heated to slightly higher than glass transition temperature T_g, only less than 20% molecules are crystallites, and more than 80% molecules are noncrystalline and
disordered. In regime II, the HST is higher than $T_g$ but lower than $T_{\text{max}}$, most molecules are crystallites, and $G$ is several orders larger than $1/t$. In regime III, the HST is larger than $T_{\text{max}}$ but lower than $T_m$, so $1/t$ is larger than $G$.

![Diagram of PET fiber model](image)

**Figure 2.6:** Gohil [110], Schematic representation of PET fiber model (a), and structure situation in the various temperature regimes for the oriented PET film (b), L represents long spacings.

Based on the paper of Gohil [110], the glass transition temperature $T_g$ is at just above 100 °C, so we see this as the big change that occurs right at 111 °C in Table 2.4.
the Tg the PET film is able to uniaxially stretch as a result of the added weight. This leads to an enhancement of the cracks (effectively increasing the crack width and increasing the resistivity). This continues over a narrow temperature range from 111 °C to 117 °C. However, as the temperature increases larger than 120 °C in region II, we see an increase in crystallinity and shrinkage of the film, which leads to the steady decrease in the $R_{AB}/R_{CD}$ Ratio. This is because a narrowing of the cracks and a large decrease in the resistance.

In summary, four different methods have been tried to improve $\frac{R_{AB}}{R_{CD}}$ of cracked ITO on PET substrate, the results are summarized in Table 2.5. The best solution among them is “Heating and Loading” process, because it keeps $R_{CD}$ staying at the lowest level (i.e. 0.35k ohm), and achieves the maximum value of $\frac{R_{AB}}{R_{CD}}$ (i.e. 33~48). We also find that the method of “Resting 24 hours” is a very good solution, which is outside the box of four initially designed methods, it keeps the value of $R_{CD}$ not changed at all, and meanwhile increases the value of $R_{AB}$ to be 3 times larger.

For the “Resting 24 hours” process which can increase the value of $R_{AB}$, we think about the heat stabilized PET substrate. The ITO PET film used in our experiment is a commercialized product, in which PET substrate is heat stabilized. Comparing with plain PET, the heat-stabilized PET is more resistant to heat, and provides improved adhesive strength and gas barrier ability for the layer coated on it [59]. That means the PET film is biaxially stretched, and then heat stabilized in a tenter with low tension applied, so the
molecule relaxation is allowed and the tendency of PET film to shrink at high temperature is reduced, e.g. shrinkage < 0.1% in air at 150 ºC 30 minutes [60]. The heat stabilized process makes the PET film to be more solid and do not shrink at high temperature (i.e. 150 ºC). Our “Resting 24 hours” process is conducted at room temperature (i.e. 25 ºC), and the ITO side on PET substrate is “elongated” because $R_{AB}$ becomes 3 times larger. So for the PET film which is not heat stabilized, we believe the “Resting 24 hours” process will make it “more elongated” because it is not as solid as the heat stabilized one.

What is happening when the ITO PET film is relaxing during the “Resting 24 hours” after it is bent can be analysis as follows. Based on the previous studies discussed in section 1.4, we know that the molecule aligned orientation and the strain-induced crystallization are the root causes of the strain hardening property of PET after it is stretched and heated. The molecule distribution is a key parameter. Therefore, we can reasonably expect that the molecule distribution and the crystallization status are changed because of the bending process. More specifically, the outer bending area is stretched, and the inner bending area is squeezed; so after the bending process, the changed molecule distribution and the changed crystallization status in these two areas begin to relax in the next 24 hours; and the final result of PET film elongation at the ITO side which increases the value of $R_{AB}$ to be 3 times larger is the superposition of the final molecule distribution and crystallization status resulted in these two areas.
The maximum value 80~33 of $\frac{R_{AB}}{R_{CD}}$ occurs at “Etching by 2.5 wt % HCl 8 minutes and Resting for 24 hours” in Method 3, but $R_{AB}$ **80k~100k ohm is not large enough** for display and touch panel applications; although $\frac{R_{AB}}{R_{CD}}$ approaches 100~66 at “Etching by 2.5 wt % HCl 14 minutes and Resting for 24 hours” in Method 3, but $R_{CD}$ **10k~30k ohm is too large**. We need to find out a better solution, which can achieve the value of $\frac{R_{AB}}{R_{CD}}$ to be several $10^2$, and keep $R_{CD}$ as small as possible, for instance, around 1k ohm.

<table>
<thead>
<tr>
<th>Method 1: Manually Stretching</th>
<th>Before Cracking</th>
<th>100</th>
<th>100</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>After Cracking</td>
<td>3k</td>
<td>0.6k</td>
<td>5</td>
</tr>
<tr>
<td>Method 2: Manually Bending at Tight Radius</td>
<td>Before Cracking</td>
<td>100</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>After Cracking</td>
<td>8k</td>
<td>0.8k</td>
<td>10</td>
</tr>
<tr>
<td>Method 3: Etching by HCl at Curved Status</td>
<td>Before Cracking</td>
<td>100</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>After Cracking and 80~100k</td>
<td>1~3k</td>
<td>80~33</td>
<td></td>
</tr>
<tr>
<td></td>
<td>“Etching 8 minutes and resting 24 hours” process</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>After Cracking and 1000~2000k</td>
<td>10~30k</td>
<td>100~66</td>
<td></td>
</tr>
<tr>
<td></td>
<td>“Etching 14 minutes and resting 24 hours” process</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Method 4: Heating and Loading</td>
<td>Before Cracking</td>
<td>100</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>After Cracking and 11~16k</td>
<td>0.35k</td>
<td>33~48</td>
<td></td>
</tr>
<tr>
<td></td>
<td>“Heating to 111~114°C and Loading 429 grams metal” process</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 2.5: The comparison of four methods tried to increase the value of $\frac{R_{AB}}{R_{CD}}$.

Method 1: manually stretching two sides of ITO on PET substrate. Method 2: the ITO on PET substrate is bent as a curvature of tight radius, and two glass plates are manually rolled to make parallel ITO cracks on PET substrate. Method 3: after making parallel ITO cracks by using method 2, the sample is immersed into 2.5 wt % HCl and etched for a period of time. Method 4: after making parallel ITO cracks by using method 2, the sample is heated in a heat oven and hung by a 429 gram metal block.
2.3 Solution of Cracking ITO: Stretching Process

After comparing the resulting performance and process stability of four methods we tried, method 2 “Tight Radius Bending” and method 4 “Heating and Loading” are selected for further improvement. The rolling process in method 2 is conducted manually, it should be more precise and reproducible, for instance, an apparatus designed to provide parallel movement during the rolling process is a good idea. The loading in method 4 is 429 gram, which can be much more strengthened if advanced equipment is applied. Therefore, the stretching process is developed in this section, which is based on the results studied in section 2.2.

As shown in Figure 2.7, we designed an apparatus consisted of two parallel rails. This apparatus was an improved design based on the idea of “Tight Radius Bending” in Figure 2.3, two improvements were the gap between two plates of glass can be precisely controlled, and two parallel rails provided precise and reproducible rolling process of making parallel ITO cracks.

The apparatus used to bend the substrates consisted of a jig to hold two rigid plates parallel with surfaces facing one another. The separation between two plates could be precisely controlled by adjusting four thumb screws. The ITO on PET film was rolled between the two plates with one end of the film attached to the top plate and the other attached to the bottom. The plate separation determined the radius of curvature. Moving one plate relative to the other rolled the ITO on PET film between two plates producing
uniformly parallel cracks in the ITO along the length of the film. The ITO on PET film was rolled five to ten times.

Figure 2.7: An apparatus consisted of two parallel rails which provide precise and reproducible rolling process of making parallel cracked ITO on PET substrate. (a) Side view. (b) Top view.

Table 2.6 shows scanning electron microscope (SEM) images of the parallel ITO cracks formed on PET substrate. As noted in previous studies, uniform bending produced uniformly parallel cracks running perpendicular to the stress direction. The cracks extended the entire width of the film and did not intersect. Resistance measured
parallel to the cracks (Rpara) is much smaller than the resistance measured perpendicular to cracks (Rperp). This is what we are looking for, because uniaxially cracked ITO stripes may be able to replace ITO electrode stripes in passive matrix displays or touch panels if the value of Rperp/Rpara is large enough.

The dependence of Rperp/Rpara was measured for different separations between two plates, as shown in Table 2.6. As expected Rperp/Rpara increased as the separation between two plates decreased. The optical microscope (OM) and SEM images of surface morphology are also included in Table 2.6. It is interesting to note that the distance between cracks (e.g. 5~10 microns) is independent of the radius of curvature in the range of below 5 mm.

<table>
<thead>
<tr>
<th>Sample #1</th>
<th>Optical Microscope (OM)</th>
<th>Scanning Electron Microscope (SEM)</th>
<th>$\frac{R_{\text{perp}}(\Omega)}{R_{\text{para}}(\Omega)}$ (ps 3 cm between probes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(In-between two plates was two ITO/PET films)</td>
<td><img src="image1" alt="Optical Microscope Image" /> <img src="image2" alt="Scanning Electron Microscope Image" /></td>
<td><img src="image3" alt="Optical Microscope Image" /> <img src="image4" alt="Scanning Electron Microscope Image" /></td>
<td>$14k / 0.7k = 20$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample #2</th>
<th>Optical Microscope (OM)</th>
<th>Scanning Electron Microscope (SEM)</th>
<th>$\frac{R_{\text{perp}}(\Omega)}{R_{\text{para}}(\Omega)}$ (ps 3 cm between probes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Separation between two plates was 2 mm)</td>
<td><img src="image5" alt="Optical Microscope Image" /> <img src="image6" alt="Scanning Electron Microscope Image" /></td>
<td><img src="image7" alt="Optical Microscope Image" /> <img src="image8" alt="Scanning Electron Microscope Image" /></td>
<td>$2.1k / 0.3k = 7$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample #3</th>
<th>Optical Microscope (OM)</th>
<th>Scanning Electron Microscope (SEM)</th>
<th>$\frac{R_{\text{perp}}(\Omega)}{R_{\text{para}}(\Omega)}$ (ps 3 cm between probes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Separation between two plates was 5 mm)</td>
<td><img src="image9" alt="Optical Microscope Image" /> <img src="image10" alt="Scanning Electron Microscope Image" /></td>
<td><img src="image11" alt="Optical Microscope Image" /> <img src="image12" alt="Scanning Electron Microscope Image" /></td>
<td>$3.5k / 0.7k = 5$</td>
</tr>
</tbody>
</table>
Table 2.6: Different values of R<sub>perp</sub>/R<sub>para</sub> were caused by different radiiuses of bending curvature (i.e. different separations between two plates). Corresponding OM and SEM pictures were also included.

The cracks were formed by the bending induced stress. The detailed root cause analysis of ITO cracks on PET substrate will be analyzed in Chapter 4. As shown in Figure 2.8, bending of the film requires an elongation of the outer surface and/or a contraction of the inner surface. Because T<sub>2</sub> << T<sub>1</sub> << R, the bending stress is mainly controlled by the thickness of PET substrate (T<sub>1</sub>) and the bending radius (R, which was half of the separation between two plates). For the same R, more stress is created by using a thicker substrate (T<sub>1</sub>). For the same substrate thickness, the stress is increased by reducing the bending radius (R).

![Diagram of ITO and PET layers](image)

**Figure 2.8:** Cracks were formed by the bending induced stress, which is the root cause of R<sub>perp</sub>/R<sub>para</sub>.

R<sub>perp</sub>/R<sub>para</sub> was also determined by the parallel uniformity of uniaxial cracks. During the process of making uniaxially cracked ITO, intersection between two neighboring cracks can occur, if the top plate is not precisely shear-stressed along two
parallel rails, as shown in Figure 2.9. After counting the number of intersections, we observed that the number of intersections in one cracked ITO stripe, in which the width was 5~10 microns and the length was 4 cm, was 3~5 in average. This number can be greatly reduced or eliminated by more precisely controlling the rolling direction and avoiding any “wobble” during the rolling process.

Figure 2.9: Intersection between two neighboring cracks can occur, if the top plate is not precisely shear-stressed along two parallel rails.

Besides extension stress, uniaxially cracked ITO could also be prepared by compression stress, as shown in Figure 2.10. ITO on PET film was first bent with the ITO on the outer surface (i.e. extension stress). The film was then turned over so the ITO was on the inner surface and bent again (i.e. compression stress). As a result, two kinds of cracks appeared in the ITO, thinner cracks were caused by extension stress, and thicker ones were caused by compression stress. It appears that the ITO delaminates from the PET substrate forming ridges in the cracks to relieve the compression stress.
Beside uniaxially cracked ITO, biaxially cracked ITO was also prepared, as shown in Figure 2.11. Isolated ITO squared islands were formed by bending the ITO on PET film along two orthogonal directions. The isolated ITO squared islands could be connected by conducting polymer offering the potential to combine the flexibility of polymer with the high conductivity of ITO [61].
The performance of uniaxially cracked ITO will be improved by increasing the differential resistance $R_{\text{perp}}/R_{\text{para}}$ and more specifically by increasing the resistance $R_{\text{perp}}$ measured perpendicular to the cracks. As noted by Crawford [62], electrical conduction across cracks is the result of contact between ITO stripes as shown in Figure 2.12. This contact between ITO stripes leads to spreading of an applied electric field beyond the contacted electrodes, effectively reducing the edge definition and resolution of a line image. We therefore explore uniaxially stretching to reduce and eliminate ITO contact across cracks.

![Contact between ITO stripes leads to spreading of an applied electric field.](image)

Figure 2.12: Contact between ITO stripes leads to spreading of an applied electric field.

In order to realize uniaxially stretching, we cooperate with Professor Mukerrem Cakmak research group in the University of Akron. Uniaxially stretching of uniaxially cracked ITO on PET film was performed at temperature 100°C with stretching loading 60~80 kg in a highly instrumented stretching system, as shown in Figure 2.13. It allowed the real-time monitoring of true stress, strain, birefringence and surface resistivity during stretching. The dumbbell shape ITO on PET film was stretched to a ratio of 1.075 with a
stretching rate 5 mm/min at 100°C. After the temperature was cooled down to room temperature, the film was released and removed from the machine. \( \frac{R_{\text{perp}}}{R_{\text{para}}} \) was improved from 10~20 to 100~300 by uniaxially stretching, and the separation between cracked ITO stripes was increased to 50 nm, as shown in Figure 2.14.

Figure 2.13: Uniaxially stretching system in Professor Mukerrem Cakmak research group in the University of Akron.

Figure 2.14: After uniaxially stretching, the separation between cracked ITO stripes is increased to 50 nm.

The geometric dimension of uniaxially cracked ITO on PET film and the direction of uniaxially stretching are shown in Figure 2.15. This dumbbell shape is very important.
Stretching of a rectangle shape film results in a compression and decreasing the width in the middle region of the film, it will produce compression cracks perpendicular to parallel cracks the stretching is meant to enhance, as shown in Figure 2.16. These compression cracks will increase $R_{\text{para}}$ and therefore decrease $R_{\text{perp}}/R_{\text{para}}$. The dumbbell shape can reduce or eliminate this compression, preventing the unwanted perpendicular cracks.

![Diagram of stress and cracks](image)

**Figure 2.15:** For the uniaxially stretching process, dumbbell shape ITO on PET film is designed to reduce the compression in the middle region.

![Image of rectangle shape ITO on PET film](image)

**Figure 2.16:** For the uniaxially stretching process, rectangle shape ITO on PET film produces compression cracks perpendicular to parallel cracks the stretching is meant to enhance.
2.4 Device: PDLC and ChLC

After preparing cracked ITO on PET substrate, the next step is to use it to make a display. Based on our expertise, the first display we make by using cracked ITO is Polymer Dispersed Liquid Crystal (PDLC) display, as introduced in Figure 1.6.

Nowadays the major application of PDLC is window product, on which the PDLC film is laminated, and the outdoor light passing through the PDLC film can be electrically controlled to be turned on (i.e. transparent) or turned off (i.e. opaque white), therefore the indoor light and the personal privacy can be adjusted by electrically switching the PDLC window. However, for all the PDLC windows in the market currently, they can only be switched for the whole display area, they cannot be selectively switched as a window curtain or a window blind, and therefore the applications of PDLC windows are limited by the shortage of functionalities.

Here we make a small area (3 cm × 3 cm) PDLC device in which one cracked ITO on PET substrate is used, as shown in Figure 2.17. The cracked ITO used in this device is made by the stretching process discussed in section 2.3, and the stretching machine in University of Akron can only stretch a sample of 3 cm × 3 cm area size. But still the possibility of selectively switching like a window curtain or a window blind is demonstrated on this device and a very unique behavior of “frequency dependent image area” can also be observed. The width of the line image driven by applying voltage of 20k Hz and 40 volts is around 5 mm, but it shrink to 1 mm by applying voltage of 200k
Hz and 40 volts. This phenomenon is very unique: the image area driven by an applied voltage can be controlled by the frequency of applied voltage.

We bonded a flexible printed circuit with an electrode width of 380 micron and electrodes separation of 190 micron to the cracked ITO substrate using ACF bonding. Individual stripes are defined by the area where the flexible printed circuit contacts the cracked ITO substrate. A clear line can be produced by charging one of the individual pads of the printed circuit.

Because of the finite resistance measured perpendicular to the cracks, adjacent ITO stripes are charged to an extent depending on the addressing frequency. The broadening of the line image is the result of bleeding of the applied electric field into adjacent ITO stripes. It is clear that the achievable resolution depends on the perpendicular resistance of the cracked ITO substrates. The resolution of line image will be maximized when the perpendicular resistance is maximized.
Figure 2.17: The structure (a) and the real device (b) of a small area (3 cm × 3 cm) PDLC device made by a cracked ITO on bottom PET substrate and a continuous ITO on top PET substrate. (b-1) Two wider 5mm lines addressed by 20k Hz and 40 volts sine wave. (b-2) Two thinner 1mm lines addressed by 200k Hz and 40 volts sine wave.
Based on our expertise, the second display we make by using cracked ITO on PET substrate is Cholesteric Liquid Crystal (ChLC) display, and its device structure and bistable behavior are illustrated in Figure 1.7.

In order to prove that the individual pixel can be displayed by using cracked ITO on PET substrate, we make a small area 3 cm × 3 cm, 7 × 7 pixels, 2 mm × 2 mm pixel size, passive matrix (PM) ChLC display in which two cracked ITO on PET substrates are used. ITO stripes on two substrates are orthogonally facing with each other, the structure and real device of PM ChLC display is illustrated in Figure 2.18. The cracked ITO used in this display is made by the stretching process in which the displayed area of substrate is 3 cm × 3 cm limited by the design of stretching machine. But still the functionality of selectively switching of a pixel image in this PM ChLC display is successfully demonstrated: Without applying electric field, 7 individual 2 mm × 2 mm green pixels of planar state and background black pixels of focal conic state are displayed, which is a clear and successful demonstration to prove the ability of “driving individual pixels” by utilizing cracked ITO stripes on PET substrate in the display.

Based on the result from abovementioned frequency-controlled line image in PDLC device, we can expect that the frequency used to drive this PM display should be high enough, because a small pixel size is what we are looking for in a high resolution display. In this 2 mm × 2 mm pixel size display, the frequency of the applied voltage is 500 Hz, the amplitude of applied voltage is 80 volts, and the cell gap is 15 microns. There is one significant result we can conclude from this PM ChLC display: Our cracked
ITO on PET substrate is useful not only in the line image window application, but also in the dot (pixel) image display application. The high resolution image is possible in the display made by cracked ITO on PET substrate, as long as the driving frequency is high enough and the width of the contact electrode is narrow enough.

The functionality over cracked ITO therefore has the potential to replace photolithography, printing, or laser etching as a simple and low cost method of forming electrodes for passive matrix displays.
Figure 2.18: The structure (a) and real device (b) of display area $3 \text{ cm} \times 3 \text{ cm}$, $7 \times 7$ pixels, pixel size $2 \text{ mm} \times 2 \text{ mm}$, PM ChLC display in which two pieces of cracked ITO on PET substrate are orthogonally assembled. The frequency of applied voltage is 500 Hz, the amplitude of applied voltage is 80 volts, and the cell gap is 15 microns.

At last, there is one unique advantage of the cracked ITO on PET substrate we should also pay attention to: The alignment-free connection between the cracked ITO stripes and the electrodes of the diving system. No matters in the PDLC window or in the ChLC display, the cracked ITO stripes are connected with the adhesive copper electrodes of the driving system directly! It means that we do not need the alignment process in which the positions of ITO electrodes need to be identified and aligned with the contact electrodes from driving system during the traditional connection process. As shown in Figure 2.9, the dimension of cracked ITO stripes is around 10 microns, and as shown in Figure 2.14, the dimension of the cracks is around 0.1 micron. Based on these numbers, we realize why it is alignment-free for the cracked ITO on PET substrate, because the ITO stripes and cracks are narrow enough. For a standard PM display, in which the dimension of pixel size is $200 \mu \text{m} \times 200 \mu \text{m}$, and the space between pixels is $30 \mu \text{m}$, there will be around 20 cracked ITO stripes included in the contact electrode of driving one pixel, and around 3 cracks included in the space between neighboring contact electrodes.

In the next chapter, a better solution of making parallel cracked ITO on PET substrate will be reported by using multiple cracking processes. This method is a cost effective solution because the chemical wet etching process is not required. It also does
not need to use the stretching machine, and therefore the size or area of cracked ITO on PET can be extended to several m², which is very important for the application of large area products, for instance, PDLC windows or touch panels.
CHAPTER 3

Parallel Cracked ITO Made by Multiple Cracking Processes

3.1 Issue of Stretching Process

After trying 5 different manufacturing processes making parallel cracked ITO on PET substrate, the rolling process conducted by an apparatus of two parallel rails (Figure 2.7) followed by the stretching process conducted by a stretching machine (Figure 2.13) has been proven to be the best solution among them. Unfortunately, this approach can only provide a sample size of 3 cm × 3 cm (Figure 2.15), which is too small in display or window application. We need a better solution to make a larger size sample which is several 10 cm × 10 cm for display application and several 50 cm × 50 cm for window application. The straightforward solution is to make a stretching machine which can stretch a larger size sample, but it will need at least hundred thousand dollars for a tailored design stretching machine we can’t afford. We need to figure out a solution which can offer the same level of $R_{perp}/R_{para}$ (100 ~ 300) achieved by the stretching process, and at the same time it have to cost within thousand dollars we can afford.

3.2 Solution of Stretching Process: Multiple Cracking Processes

In this section a technology is invented to enhance the cracks in ITO. As noted above the cracks in ITO are produced by bending PET substrate around a tight radius of curvature with ITO being an outer layer of the bending. This leads to uniform cracks that extend uniformly across the width of the film. Also as noted above when the film is
returned to the flat orientation, neighboring ITO touches across the crack, resulting in a relatively low value of the differential resistance.

We find that the cracks can be enhanced by rolling the film multiple times: First outwardly bending, and inwardly bending, as shown in Figure 3.1(a). The location of each crack is defined during the first rolling step, and additional rolling steps serve to enhance the cracks. We observe delamination of ITO layer near the cracks. Microscopic images of the ITO cracks formed after the first bending then followed by multiple bending steps which clearly enhance the cracks are shown in Figure 3.1. Multiple bending steps greatly increase the resistance ratio to about 200. This is the same order of magnitude as achieved through uniaxial stretching process performed by the stretching machine in the University of Akron.

(a)
Figure 3.1: (a) Comparison of cracking ITO processes, (a, left) Previous method, first the ITO PET film is outwardly bent while ITO layer is on the outside, and then it is stretched, (a, right) Improved method, first the ITO PET film is outwardly bent while ITO layer is on the outside, and then it is inwardly bent while ITO layer is on the inside. (b) Microscopic images of improved cracked ITO on PET substrate, (b, left) After the first outwardly bending process, (b, right) After the inwardly bending process.

This multiple bending approach simplifies the manufacturing process, eliminating the chemicals required for soft etching or the heat and controlled elongation process required for uniaxial stretching. This multiple rolling process can easily be incorporated into a roll-to-roll manufacturing line to produce the uni-axially cracked ITO on PET substrate.

The efficacy of this multiple cracking technology is demonstrated by producing a selectively switchable polymer dispersed liquid crystal (PDLC) window shutter. They consist of droplets of liquid crystal dispersed in a thin polymer film. In the absence of applied electric field, the directors of liquid crystal droplets are randomly aligned
resulting in a significant mismatch of the refractive index of droplet and the surrounding polymer binder. In this state the film efficiently scatters light and they appear white. Application of an external electric field across the PDLC film serves to align the liquid crystal directors parallel to the field. The polymer binder is selected to match the refractive index of the aligned liquid crystal droplets and the film turns clear. PDLC have found their most commercial success for electrically switchable windows. These windows are made using continuous ITO layer coated on a flexible PET substrate. A thin film of PDLC is sandwiched in between two of these substrates. Using the continuous ITO layers, the PDLC film is either entirely in the OFF or ON state.

3.3 Prototype of PDLC Window Shutter having Tunable Function

We make a selectively addressed PDLC window using one cracked ITO substrate and one continuous ITO substrate. We again use narrow 2 mm width stripes of adhesive copper tape to contact and define the electrodes on the cracked ITO substrate. We design a custom drive scheme to selectively address the individual electrodes and to vary the applied voltage. Because of the residual conductivity across the cracks, the ITO electrodes adjacent to the directly contacted copper tape will be charged to a certain level that depends on the distance between the cracked ITO stripe and the contacted copper tape, the magnitude and the frequency of applied field. The details of electrical property of the cracked ITO and how the characteristics of applied electric field may be tailored to control the effective width of the addressed ITO stripes are studied in Chapter 5. It is clear that the width of addressed line is increased as the frequency of the applied electric
field is decreased. This is likely explained by the RC time constant in the equivalent circuit. The results will be reported in Chapter 6.

The major application of PDLC is window, but the display area of device we made in Chapter 2 was very small $3 \text{ cm} \times 3 \text{ cm}$ (Figure 2.15), because of the limitation of stretching machine. Although the frequency-controlled line width of driving image have been demonstrated in that device, it is still not a direct evidence claiming that the PDLC window made by our cracked ITO on PET substrate can work in a real window size product which has the tunable function required for window shutter and window curtain applications.

Because of the improvements described in Chapter 3, we are allowed to make a real window size ($26 \text{ cm} \times 42 \text{ cm}$) PDLC window in which one real window size cracked ITO on PET substrate is used, as shown in Figure 3.2. The cracked ITO used in this device is made by the multiple times bending process which free us to achieve large area of the substrate. Large area is very important for the window application, because we cannot convince customers to believe our cracked ITO technology is useful in window application, if there is only a $3 \text{ cm} \times 3 \text{ cm}$ device demonstrated.
Figure 3.2: A real window size (26 cm × 42 cm) PDLC window. (a) Off state. (b) On state, applied with 60 volts 50 Hz for the whole display area. (c) The contact electrodes from driving system are attached on cracked ITO stripes.

As shown in Figure 3.2 (c), there are only 14 pairs (i.e. 28 contacts) effective electrodes are connected to the cracked ITO stripes on PET substrate. The driving voltages are provided by the driving system (i.e. white box), which controls the amplitude and frequency of output voltage, and also determines which electrode will generate output voltage.

The importance of the multiple times bending process is much clearer now. Because theoretically it is possible to develop a stretching machine which allows us to
stretch a large size cracked ITO on PET film; but practically this is not feasible, because it will need much more funding we can’t afford. If we did not figure out and implement this simple, cost-effective and reproducible solution of multiple times bending process, then we will never have a real large area PDLC window made by the cracked ITO on PET substrate.

Next, with the help of electronic driving system, we implemented the selectively switching function of this large area PDLC window, and make it look like a traditional window curtain and window shutter. Meanwhile, the unique behavior of the frequency dependent line width of driving image can also be directly used to provide the unique “tunable window shutter” or “venetian blind” function which can be achieved only by using the cracked ITO on PET substrate, because one contacted copper tape electrode can control the neighboring cracked ITO stripes by applying different frequency of applied electric field.

The multiple functions of PDLC window made by the cracked ITO on PET substrate are described as follows.

First, the “window curtain” function, i.e. the “drawable” function, as shown in Figure 3.3, 60 volts 50 Hz external voltage is applied on selected electrodes in transparent area. The microcontroller in driving system can select which one of contacted adhesive copper electrode is applied by external voltage, so we can choose the transparent area. For instance, the bottom area is choosen to be transparent as shown in Figure 3.3 (a), and the middle and bottom areas are choosen to be opaque white as shown in Figure 3.3 (b).
Figure 3.3: The “window curtain” function of PDLC window. (a) Drawing to reveal the bottom area of the window. (b) Drawing to reveal the middle and bottom areas of the window. 60 volts 50 Hz external voltage is applied on selected electrodes in the transparent area.

Second, the “window shutter” or “venetian blind” function, i.e. “tunable” function, as shown in Figure 3.4, 60 volts external voltage of different frequencies is
applied on all electrodes in the transparent area. The microcontroller in driving system can set the frequency of external voltage, so as the line width of driven transparent area. For instance, 2000 Hz is set to drive a wider 3 cm line width as shown in Figure 3.3 (a), and 5000 Hz is set to drive a narrower 3 mm line width as shown in Figure 3.3 (b).

Figure 3.4: The “window shutter” or “venetian blind” function of PDLC window. (a) Tuning to reveal wider 3 cm transparent lines, 60 volts 2000 Hz voltage is applied.
on all electrodes. (b) Tuning to reveal narrower 3 mm transparent lines, 60 volts 5000 Hz is applied on all electrodes.

Third, the combination of drawable and tunable functions, as shown in Figure 3.5, 60 volts external voltage of different frequencies is applied on selected electrodes in transparent area. The microcontroller in driving system can select which one of contacted adhesive copper electrode is applied by setting the specific frequency of external voltage, so we can choose the transparent area and the selected line width of driving image. For instance, 2000 Hz is selected to drive a wider 3 cm line width of transparent area in the bottom area as shown in Figure 3.5 (a), and the middle and bottom areas as shown in Figure 3.5 (b); 5000 Hz is selected to drive a narrower 3 mm line width of transparent area in the bottom area as shown in Figure 3.5 (c), and and the middle and bottom areas as shown in Figure 3.5 (d).
Figure 3.5: The curtain and shutter functions of PDLC window. (a)~(b) Drawing to reveal the bottom area (a) or middle and bottom areas (b) of the window, and tuning to reveal wider 3 cm transparent lines, 60 volts 2000 Hz voltage is applied on selected electrodes. (c)~(d) Drawing to reveal the bottom area (c) or middle and bottom areas (d) of the window, and tuning to reveal narrower 3 mm transparent lines, 60 volts 5000 Hz voltage is applied on selected electrodes.
CHAPTER 4
Physics of Cracked ITO on PET Substrate

As demonstrated in Chapter 2 and Chapter 3, plane ITO on PET substrate can be well controlled to generate uniform and long lasting parallel cracks which are useful in making devices and products of PDLC, ChLC, and window shutter. The next question we want to ask is: why these uniform and long lasting parallel cracks can be generated? We know that a sheet of glass plate will become broken pieces if we roll it up. Why the cracks on PET substrate are so uniform? Does it relate to crystalline or amorphous structure of ITO or PET? What is the physics inside this phenomenon? In order to answer these questions, the physics of solid mechanics need to be studied. Beuth [63] had provided a quantitative analysis for two kinds of cracked thin film (i.e. fully cracked thin film and partially cracked thin film) bonded on a thick substrate in 1992. Based on Beuth’s result, Xia [64] had developed a two-dimensional model for simulating crack propagation paths of a thin film bonded on a thick substrate in 2000. Based on the achievements completed by Beuth [63] and Xia [64], our experimental results of cracked ITO on PET substrate are analyzed in this chapter.

4.1 Definition of Solid Mechanics

In order to have a clear understanding of the basic concepts in solid mechanic, several important definitions are reviewed as follows.
• **Stress** [65–67] is defined as the average force per unit area that some particles of a body exerts on adjacent particles, across an imaginary surface that separates them, as shown in Figure 4.1 and Figure 4.2 (a).

![Stress](image)

Figure 4.1: Stolfi [67], a picture to illustrate the definition of stress. The force applied by a body (top sphere) on another body (bottom sphere) through the connection surface (yellow disk) that separates them is represented by the large purple arrow. The stress (the smaller arrows distributed on the yellow disk) is the force divided by the area of the connection surface.

• **Tension** [68] is the pulling force exerted by each end of a string, cable, chain, or similar continuous body. Tension is the opposite of compression [69], tension is when the forces acting on a body are trying to stretch it, but compression is when the forces acting on a body are trying to squash it.

• **Tensile** [69] means a body is under Tension.

• **Tensile Stress** is the stress in a body which is under Tension.
• **Tensor** [70] is a geometric object that describes linear relations between vectors, scalars, and other tensors. Elementary examples of such relations include the dot product, the cross product, and linear maps. For example, as a vector with respect to a given basis is represented by an array of one dimension, any tensor with respect to a basis is represented by a multidimensional array.

• **Stress Tensor** [71] is a $3 \times 3$ matrix of stress components, as shown in Figure 4.2 (b).

![Stress Tensor Diagram](image)

Figure 4.2: eFunda Inc. [72], Definition of stress (a) and stress tensor (b).
• **Deformation** [73] is the transformation of a body from a reference configuration to a current configuration. A configuration is a set containing the positions of all particles of the body, as shown in Figure 4.3.

• **Strain** [78] is a normalized measure of deformation representing the displacement between particles in the body relative to a reference length, as shown in Figure 4.3 and 4.4.

![Diagram of deformation and strain](image)

**Figure 4.3:** Mircalla22 [74], An example of calculating the strain of the deformation of a rectangle in x y plane.

• **Young’s modulus** $E$, or **Tensile modulus**, or **Elastic modulus** [76] is a measure of the stiffness of an elastic material and is a quantity used to characterize materials, it is defined as the ratio of the stress (force per unit area) along an axis to the strain (ratio of deformation over initial length) along that axis in the range of stress in which Hooke's law holds, as shown in Figure 4.4.
• **Poisson’s ratio** $\nu$ [78] is the negative ratio of vertical strain to lateral strain (i.e. two orthogonal strains), it is the fraction of expansion (or compression) divided by the fraction of compression (or expansion), for small values of these changes, as shown in Figure 4.4.

![Figure 4.4](image)

Figure 4.4: Batzle [77], the definition of vertical strain, Young’s modulus, lateral strain, and Poisson’s ratio.

• **Shear modulus** $\mu$ [79] is the ratio of shear stress to shear strain, as shown in Figure 4.5.
Figure 4.5: The definition of shear stress, shear strain, and shear modulus.
4.2 Previous Study of Cracks in Film bonded by Substrate

Figure 4.6: Beuth [63], an isotropic film (labeled 1) having thickness $h$ is bonded on an isotropic substrate (labeled 2), when stress $\sigma$ is applied on the film, the steady state of cracked film will be: (a) fully cracked film, in which a single crack extending down to the film/substrate interface is generated, or (b) partially cracked film, in which a single crack extending down to the depth of $a$ in the film is generated. (c) Perspective picture of (a).

Beuth [63] had provided a quantitative analysis for cracking of thin bonded films in 1992. As shown in Figure 4.6, two kinds of cracked film can be generated in an isotropic film bonded to an isotropic substrate, when stress $\sigma$ is applied on the film: a
steady state, (a) fully cracked film, or (b) partially cracked film, will be the result. The perspective picture of fully cracked film is illustrated in (c).

In our study, there are parallel cracks in ITO film on PET substrate. After the first bending process, slightly cracked parallel cracks are formed, but two neighboring ITO stripes beside the same crack are still touch with each other, as shown in Figure 3.1 (left); therefore it looks like a result of partially cracked film. After that, the second bending process is continuously performed, and two neighboring ITO stripes beside the same crack are mostly separated, as shown in Figure 3.1 (right). Therefore, our cracked ITO on PET substrate is much more complicated than Beuth’s extension stress model, because our sample is formed by extension and compression stresses. But still we can study Beuth’s model first to get a partial picture of what happens in our cracked ITO on PET substrate. In order to use Beuth [63], we need to know the following parameters.

Two dimensionless quantities are defined as

$$F(\alpha, \beta, \frac{a}{h}) = \frac{K_1}{\sigma \cdot (\pi h)^{1/2}}$$

$$G(\alpha, \beta, \frac{a}{h}) = \frac{\int \delta(y) \cdot dy}{\pi \cdot \sigma \cdot a \cdot h}$$

where

$$\alpha = \frac{E - E_s}{E + E_s} \quad \text{and} \quad \beta = \frac{1}{2} \frac{\mu \cdot (1 - 2\nu_s) - \mu_s \cdot (1 - 2\nu)}{\mu \cdot (1 - \nu_s) + \mu_s \cdot (1 - \nu)}$$

where
\[
\bar{E} = \frac{E}{1 - \nu^2}, \quad \text{is Young’s modulus of the film, } \nu \text{ is Poisson ratio of the film}
\]

\[
\bar{E}_s = \frac{E_s}{1 - \nu_s^2}, \quad \text{is Young’s modulus of the substrate, } \nu_s \text{ is Poisson ratio of the substrate}
\]

\[
\mu = \frac{E}{2(1 + \nu)} \quad \text{is the sheer modulus of the film}
\]

\[
\mu_s = \frac{E_s}{2(1 + \nu_s)} \quad \text{is the sheer modulus of the substrate}
\]

\(K_i\) is the stress intensity factor, which predicts the stress intensity near the tip of a crack caused by a remote load or residual stresses. \(\delta(y)\) is the crack opening displacement. Beuth [63] solved the Burgers vector magnitude \(b(\xi)\), and then \(K_i\) and \(\delta(y)\) are given.

Based on Beuth’s result, Xia [64] developed a model for crack patterns in fully cracked thin film on substrate in 2000. With \(\Gamma c\) as the mode-I fracture toughness of the film measured in units of energy per unit area, the condition for propagation of an isolated crack across a brittle film is

\[G = \Gamma c\]

When the pre-stress \(\sigma^0\) and/or film thickness \(h\) are small, the energy release rate \(G\) is smaller than the mode-I fracture toughness \(\Gamma c\), so there are only short cracks and crack-like flaws; when the pre-stress \(\sigma^0\) and/or film thickness \(h\) are large, the energy release rate \(G\) is larger than the mode-I fracture toughness \(\Gamma c\), so there is an isolated crack propagating across a brittle film.
Similarly, as shown in Figure 4.10, when the function $g(\alpha, \beta)$ is small, the energy release rate $G$ is small, and therefore the normalized length $\ell / h$, which defines the in-plane resistance of the film, is small; when the function $g(\alpha, \beta)$ is large, the energy release rate $G$ is large, and therefore the normalized length $\ell / h$ is large. The reference length $\ell$ which characterizes the exponential decay of the changes transverse to the crack is defined as

$$\ell \equiv \sqrt{\frac{E \cdot h}{k}} = \frac{\pi}{2} \cdot g(\alpha, \beta) \cdot h$$

where $k$ is the substrate spring constant.

**Figure 4.7:** Xia [64], for $\beta = 0$ (the dependence on $\beta$ is weak), the reference length $\ell$ which characterizes the exponential decay of the changes transverse to a steady-state fully cracked crack in a film. The normalized length $\ell / h$ which defines the in-plane resistance of the film is determined by the function $g(\alpha, \beta)$. 

80
4.3 Cracked ITO on Different Substrates

After making cracked ITO on PET substrate, we continue to make cracked ITO on different plastic substrates (e.g. PC and PEN) to compare and analyze the differences between them, which is also helpful to test the reliability of models developed by Beuth [63] and Xia [64] discussed above.

ITO coated on three different plastic substrates is provided by Genvac Aerospace Inc. as follows: ITO film on PET substrate, ITO film on PC substrate, and ITO film on PEN substrate. They are bent at 3 mm radius of curvature and rolled 6 times, which is the identical process we did on the ITO on PET provided by Sheldahl (Accentia, 125 µm, 60 ohms per square). After bending process, they are sent to the University at Akron to conduct the stretching process. The comparison of resistance ratio in these four samples is summarized in Table 4.1.

<table>
<thead>
<tr>
<th></th>
<th>Film Thickness (µm)</th>
<th>$R_{AB}(\text{ohm})/R_{CD}(\text{ohm})$ (After rolling 6 times at 3 mm radius of curvature)</th>
<th>Tg (C)</th>
<th>Stretching temp, Stretching ratio</th>
<th>Relaxing during stretching temperature</th>
<th>$R_{AB}(\text{ohm})/R_{CD}(\text{ohm})$ (After stretching)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Genvac ITO/PEN</td>
<td>170 ~ 180</td>
<td>50k/2k = 25</td>
<td>PEN ~ 120</td>
<td>140C, 5%</td>
<td>Yes (floppy)</td>
<td>300k/5k = 60 ~ 400k/5k = 80</td>
</tr>
<tr>
<td>Genvac ITO/PET</td>
<td>120 ~ 130</td>
<td>20k/1.5k = 13</td>
<td>PET ~ 70</td>
<td>120C, 5%</td>
<td>No (hold tight)</td>
<td>120k/20k = 60 ~ 65k/1.5k = 43</td>
</tr>
<tr>
<td>Genvac ITO/PC</td>
<td>190 ~ 200</td>
<td>10k/1k = 10</td>
<td>PC ~ 150</td>
<td>170C or 150C, 5%</td>
<td>Yes (floppy)</td>
<td>170C: 13k/1k = 13</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>150C: 20k/1k = 20</td>
</tr>
<tr>
<td>Sheldahl ITO/PET</td>
<td>125</td>
<td>5k/1k = 5</td>
<td>PET ~ 70</td>
<td>120C, 5%</td>
<td>No (hold tight)</td>
<td>1000k/10k = 100</td>
</tr>
</tbody>
</table>

Table 4.1: The comparisons of four samples of ITO on plastic substrate, the resistance ratio after rolling process and stretching process are summarized.
In order to compare the small scale differences of cracked ITO on different plastic substrate, we use optical microscope to observe the cracks on three Genvac samples at higher magnification. The pictures after stretching process make our comparison easier, because the cracks are much clearer after the stretching process. The comparison of microscopic pictures (1000×) for three Genvac samples after rolling process and stretching process is summarized in Table 4.2.

<table>
<thead>
<tr>
<th></th>
<th>Microscope(1000x)</th>
<th>Microscope(1000x)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(After rolling 6 times at 3 mm radius of curvature)</td>
<td>(After stretching)</td>
</tr>
<tr>
<td>Genvac ITO/PEN</td>
<td><img src="image" alt="Genvac ITO/PEN Microscope" /></td>
<td><img src="image" alt="Genvac ITO/PEN Microscope" /></td>
</tr>
<tr>
<td></td>
<td><img src="image" alt="Genvac ITO/PEN 20 Micrometer" /></td>
<td><img src="image" alt="Genvac ITO/PEN 20 Micrometer" /></td>
</tr>
<tr>
<td>Genvac ITO/PET</td>
<td><img src="image" alt="Genvac ITO/PET Microscope" /></td>
<td><img src="image" alt="Genvac ITO/PET Microscope" /></td>
</tr>
<tr>
<td></td>
<td><img src="image" alt="Genvac ITO/PET 20 Micrometer" /></td>
<td><img src="image" alt="Genvac ITO/PET 20 Micrometer" /></td>
</tr>
</tbody>
</table>
Table 4.2: The comparison of microscopic pictures (1000×) for three Genvac samples after rolling process and stretching process is summarized.

In order to compare the large scale differences of cracked ITO on different plastic substrate, we use optical microscope to observe the cracks on three Genvac samples at lower magnification. The comparison of microscopic pictures (400×, 200×, 100×) for three Genvac samples after stretching process is summarized in Table 4.3.
Table 4.3: The comparison of microscopic pictures (400×, 200×, 100×) for three Genvac samples after stretching process is summarized.

### 4.4 Explanation of Different Cracked ITO Patterns on Different Substrates

The model developed by Beuth [63] can be used to explain different patterns observed in our experiments of parallel cracked ITO on PET, PC, and PEN substrates. The values of Young’s modulus and Poisson ratio of ITO, PET, PC, and PEN are looked up from several different websites on the internet and the average of them are calculated, as shown in Table 4.4. Then, the values of $\mu$, $\overline{E}$, $\overline{E_r}$, $\alpha$, $\beta$ are calculated by using these averaged values, as shown in Table 4.4 and 4.5.
ITO film (sputtered, weight 10% SnO$_2$) & PET substrate & PC substrate & PEN substrate \\
Young’s modulus $E$ ($10^9$ N/m$^2$ = GPa) & 116 [80] & 4 [81] & 2–2.4 [84] & 5 [81] \\
& 145 [81] & 3.5 [82] & 2.4 [85] & 6.08 [87] \\
Averaged value & 130 & 3.5 & 2.2 & 5.5 \\
Poisson ratio $\nu$ & 0.35 [80] & 0.3 [81] & 0.37 [84] & 0.3–0.4 [81] \\
& 0.2 [81] & 0.39 [82] & 0.37–0.38 [86] & 0.3–0.4 [88] \\
Averaged value & 0.28 & 0.35 & 0.37 & 0.35 \\
Shear modulus $\mu$ & $\mu = \frac{E}{2(1+\nu)}$ & 50.781 & 1.296 & 0.802 & 1.852 \\
Table 4.4: The looked up values of Young’s modulus, Poisson ratio, and the calculated values of Shear modulus: ITO, PET, PC, and PEN. \\

$\alpha$ and $\beta$ are defined for the system of “the film bonded on the substrate”, so:

$$
\overline{E} = \frac{E}{1-\nu^2}
$$

<table>
<thead>
<tr>
<th>$\overline{E}$</th>
<th>ITO film on PET substrate</th>
<th>ITO film on PC substrate</th>
<th>ITO film on PEN substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>141.059</td>
<td>141.059</td>
<td>141.059</td>
<td></td>
</tr>
</tbody>
</table>

$$
\overline{E_s} = \frac{E_s}{1-\nu_s^2}
$$

<table>
<thead>
<tr>
<th>$\overline{E_s}$</th>
<th>ITO film on PET substrate</th>
<th>ITO film on PC substrate</th>
<th>ITO film on PEN substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.989</td>
<td>2.549</td>
<td>5.698</td>
<td></td>
</tr>
</tbody>
</table>

$$
\alpha = \frac{\overline{E} - \overline{E_s}}{\overline{E} + \overline{E_s}}
$$

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>ITO film on PET substrate</th>
<th>ITO film on PC substrate</th>
<th>ITO film on PEN substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.945</td>
<td>0.965</td>
<td>0.922</td>
<td></td>
</tr>
</tbody>
</table>

$$
\beta = \frac{1}{2} \frac{\mu_s(1-2\nu_s) - \mu(1-2\nu)}{\mu_s(1-\nu_s) + \mu(1-\nu)}
$$

<table>
<thead>
<tr>
<th>$\beta$</th>
<th>ITO film on PET substrate</th>
<th>ITO film on PC substrate</th>
<th>ITO film on PEN substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.216</td>
<td>0.197</td>
<td>0.210</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.5: $\overline{E}$, $\overline{E_s}$, $\alpha$, $\beta$ in three systems of “film bonded on substrate”: ITO film on PET substrate, ITO film on PC substrate, and ITO film on PEN substrate.
These calculated values of $\alpha$ and $\beta$ are fall in the expected region, which is $0 \leq \alpha \leq 1$ and $0 \leq \beta \leq \frac{\alpha}{4}$. Based on these values, the cracked ITO on plastic substrates of PEN, PC, and PEN can be explained and predicted as follows.

First, they are all fully cracked ITO film on PET or PC or PEN substrate, because $\alpha \approx 1$, i.e. $E >> E_s$, ITO film is stiff with respect to the substrate, there is no maximum of $F(\alpha, \beta, \frac{a}{h})$ occurs in $0 \leq \frac{a}{h} < 1$, and $F(\alpha, \beta, \frac{a}{h})$ approaches infinity as $\frac{a}{h}$ approaches 1. Thus, given a flaw of certain size in the stiff film, once $K_i$ is larger than $K_{ic}$, the crack will grow all the way to the interface between the film and the substrate, i.e. it is a fully cracked film.

Second, the reference length $\ell = \sqrt{\frac{E \cdot h}{k}} = \frac{\pi}{2} \cdot g(\alpha, \beta) \cdot h$, which characterize the exponential decay of the changes transverses to the crack, in “ITO film on PC substrate” should be longer than “ITO film on PET substrate” or “ITO film on PEN substrate”, because $g(\alpha, \beta)$ is 14.3883 in “ITO film on PC substrate” which is larger than 8.5809 in “ITO film on PET substrate” or 7.1290 in “ITO film on PEN substrate”, as shown in Table 4.7. The values of $f(\alpha, \beta)$ and $g(\alpha, \beta)$ are calculated by linear interpolation using the values within the red frames in Table 4.6.
Table 4.6: Beuth [63], the look-up table of $f(\alpha, \beta)$ and $g(\alpha, \beta)$ in fully cracked film.

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>$f(\alpha, 0)$</th>
<th>$f(\alpha, 1/4)$</th>
<th>$f(\alpha, 1/2)$</th>
<th>$f(\alpha, 1)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.99</td>
<td>1.780</td>
<td>1.940</td>
<td>2.200</td>
<td>2.400</td>
</tr>
<tr>
<td>0.95</td>
<td>1.760</td>
<td>1.928</td>
<td>2.180</td>
<td>2.380</td>
</tr>
<tr>
<td>0.90</td>
<td>1.720</td>
<td>1.900</td>
<td>2.140</td>
<td>2.340</td>
</tr>
</tbody>
</table>

Table 4.7: $f(\alpha, \beta)$ and $g(\alpha, \beta)$ are calculated by using linear interpolation and the values within the red frames in Table 4.6.

The reference length $l$ characterizes the exponential decay of the changes transverses to the crack, so physically it should be proportion to the width of crack, i.e. the separation between ITO stripes. By comparing the width of ITO cracks on three
different plastic substrates in Table 4.2 and 4.3, we surprisingly find that the width of ITO cracks in “ITO film on PC substrate” are indeed 3 to 5 times wider than that in “ITO film on PET substrate” or “ITO film on PEN substrate”. The prediction of reference length \( \ell \) proportioning to \( g(\alpha, \beta) \) is consistent with the observation in our experiment.

Third, different patterns of cracked ITO on different substrates are caused by different values of \( E_s = \frac{E_s}{1-\nu_s^2} \) of PET, PC, and PEN, where \( E_s \) is Young’s modulus of substrate, and \( \nu_s \) is Poisson ratio of substrate. This prediction is based on the model developed by Xia [64]. As shown in Figure 4.8, for the same value of \( \frac{\ell}{H} \), a smaller value of \( \overline{E} \) will induce the sequential process of cracks, and a larger value of \( \overline{E} \) will induce the simultaneous process of cracks. This prediction is also consistent with the observation in our experiment result, because \( \overline{E_s} \) of PC is 2.549, which is smaller than that of 3.989 in PET or 5.689 in PEN; as shown in Table 4.3, there are some “short length” cracks observed in “ITO on PC substrate” in which some cracks are not long lasting from the beginning to the end, they are very similar to the sequential process of cracks predicted in Figure 4.11; and only “long length” cracks are observed in “ITO on PET substrate” and “ITO on PEN substrate” in which cracks are long lasting from the beginning to the end, they are very similar to the simultaneous process of cracks predicted in Figure 4.8.
Another feature of the pattern of cracks is the width of ITO stripes, i.e. $H$. As shown in Figure 4.8, for the same value of $\frac{G\bar{E}}{\sigma_0^2\ell}$, a smaller value of $\frac{\ell}{H}$ will be responding to the sequential process of cracks, and a larger value of $\frac{\ell}{H}$ will be responding to the simultaneous process of cracks. A smaller $\frac{\ell}{H}$ means a larger $H$, which is again consistent with the observation in our experiment result, because the width of ITO stripes is indeed 3 to 5 times longer in the “sequential process crack pattern” in “ITO on PC substrate” than that in the “simultaneous process crack pattern” in “ITO on PET substrate” or “ITO on PEN substrate”.

Figure 4.8: Xia [64]. Energy release rate at each crack tip for steady-state channeling of parallel cracks in the film bonded on substrate. The upper curve applies to the simultaneous advance of all cracks, and the lower curve applies to the sequential process where a new set of cracks propagate midway between a previously formed set of cracks.
4.5 How to Optimize Parallel Cracked ITO on Plastic Substrate

Based on the analysis in CHAPTER4, we have following approaches to optimize the parallel cracked ITO on plastic substrates, in which the value of \((R_{\text{perp}} / R_{\text{para}})\) is maximized.

The first approach is to increase the thickness of ITO film or plastic substrate, the reason is: the deformation of ITO film or plastic substrate bending at a radius of curvature is increased, if the thickness of ITO film or plastic substrate is increased, as shown in Figure 2.8; and a larger deformation means a larger strain applied on ITO film or plastic substrate, as shown in Figure 4.4. Evidence is shown in Table 4.8, four samples of parallel cracked ITO on PET substrate having different thickness of ITO film and PET substrate are outwardly bent 6 times at 3 mm radius of curvature, and the \(R_{\text{perp}}\) and \(R_{\text{para}}\) are measured after bending. It is clear that the value of \((R_{\text{perp}} / R_{\text{para}})\) is increased as the thickness of ITO film or PET substrate is increased.

<table>
<thead>
<tr>
<th>Sheet Resistance of ITO</th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Sample 3</th>
<th>Sample 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>200 ohm/sq (Shedal)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thickness of PET Film</td>
<td>5 mil = 125 μm</td>
<td>2 mil = 50 μm</td>
<td>5 mil = 125 μm</td>
<td>2 mil = 50 μm</td>
</tr>
<tr>
<td>(R_{\text{para}}) after bending (ohm)</td>
<td>(6 ~ 8) k</td>
<td>5 k</td>
<td>(0.4 ~ 0.5)k</td>
<td>0.7 k</td>
</tr>
<tr>
<td>(R_{\text{perp}}) after bending (ohm)</td>
<td>600 k</td>
<td>(50 ~ 100) k</td>
<td>(80 ~ 100) k</td>
<td>15 k</td>
</tr>
<tr>
<td>(R_{\text{perp}} / R_{\text{para}})</td>
<td>100 ~ 75</td>
<td>10 ~ 20</td>
<td>200</td>
<td>21.5</td>
</tr>
</tbody>
</table>

Table 4.8: Four samples of parallel cracked ITO on PET substrate having different thickness of ITO film and PET substrate are outwardly bent 6 times at 3 mm radius of curvature.
The second approach is to create a plastic substrate in which Young’s Modulus, $E_s$ and $g(\alpha, \beta)$ are between these values of PET and PC. The reason is as follows. As shown in Table 4.1, the value of $(R_{AB}/R_{CD})$ in Genvac ITO on PC (i.e. 13 ~ 20) is much smaller than that in Genvac ITO on PEN (i.e. 60 ~ 80) and Genvac ITO on PET (i.e. 60 ~ 43), which means the cracked ITO on PC is much different from that on PEN and PET. This phenomenon can be observed more clearly by comparing the pictures in Table 4.2 and 4.3. The width of ITO cracks in “ITO film on PC substrate” are 3 to 5 times wider than that in “ITO film on PET substrate” or “ITO film on PEN substrate”. But, the lines of ITO cracks in “ITO film on PC substrate” are interrupted, and that in “ITO film on PET substrate” or “ITO film on PEN substrate” are continuously straight lines. So, the optimized “ITO on something of substrate” in which the something should has the property between PET and PC.

More specifically, based on the values from Table 4.4, Table 4.5, and Table 4.7, we have:

$$2.2 \text{ (PC)} < \text{Young’s Modulus of optimized plastic substrate} < 3.5 \text{ (PET)}$$

$$2.549 \text{ (PC)} < E_s \text{ of optimized plastic substrate} < 3.989 \text{ (PET)}$$

$$8.5809 \text{ (ITO on PET)} < g(\alpha, \beta) \text{ of optimized plastic substrate} < 14.3883 \text{ (ITO on PC)}$$

The optimized plastic substrate should be able to maintain the ITO cracks as continuously straight lines and meanwhile the width of ITO cracks should be wider than that in the current “ITO film on PET substrate” system.
CHAPTER 5

Frequency Dependent Line Width of Driving Image

For the PDLC window made by our cracked ITO on PET substrate, the curtain and shutter functions are demonstrated in section 3.4. The line width of driving image is controlled by the frequency of applied voltage, which is the critical issue we will analyze in the next two chapters. In this chapter, we will measure the frequency dependent behavior quantitatively. Different frequencies of the applied voltage and different lengths of the ITO cracks will be tested. The amplitude and phase of response voltage by applying different frequencies and measuring at different positions of a device made by the cracked ITO on PET substrate will be measured and plotted as well.

5.1 Frequency Independent Line Width of Conventional PDLC Device

For the PDLC window in which conventional patterned ITO stripes on PET substrate is used, the structure is shown in Figure 5.1. Conventional patterned ITO stripes are implemented by using patterning technology such as photo-lithography or laser patterning, and two neighboring ITO stripes are completely isolated. When a voltage is applied to one ITO stripe, the response voltages on the neighboring ITO stripes are 0 volt, no matter what frequency is used in the applied voltage, because there is no connection between these two ITO stripes. Therefore, the line width of the driving image is independent on the frequency of applied voltage, it is only dependent on how many connections are built between the ITO stripes and the electrodes of applied voltage.
Figure 5.1: The PDLC window in which the bottom plate is made by conventional patterned ITO stripes on PET substrate, and the top plate is made by continuous ITO plane on PET substrate. Two neighboring ITO stripes in the bottom plate are completely isolated by using the photo-lithography or laser patterning process. The line width of driving image is independent on the frequency of applied voltage, and it is only dependent on how many connections are built between the ITO stripes and the electrodes.

5.2 Different Frequencies of Applied Voltage

For the PDLC window made by using our cracked ITO on PET substrate, the structure is shown in Figure 5.2. Our cracked ITO stripes are implemented by using the bending technology mentioned in CHAPTER2 and CHAPTER3; therefore two neighboring ITO stripes are not completely isolated, there are still tiny connections between them. When a voltage is applied to the contacted cracked ITO stripes, the response voltages on the neighboring ITO stripes are not 0 volt, because we can observe that the line width of driving image, as shown in Figure 5.3, is wider than the width of...
electrode (i.e. adhesive copper tape 2 mm) which defines the width of the contacted cracked ITO stripes. The response voltage $V(x)$ at position $x$ is $V_e \cdot \sin(\omega \cdot t + \phi_e)$, in which the amplitude is changed from $V$ to $V_e$, the frequency is unchanged, and the phase is changed from 0 to $\phi_e$.

\[
V(x) = V_e \cdot \sin(\omega \cdot t + \phi_e)
\]

Figure 5.2: The PDLC window in which the bottom plate is made by our cracked ITO stripes on PET substrate, and the top palte is made by continuous ITO plane on PET substrate. Two neighboring ITO stripes in the bottom plate are still connected. The line width of driving image is dependent on the frequency of applied voltage.

Two interesting phenomena are discovered. First, the line width of driving image is dependent on the frequency of applied voltage; second, the line width of driving image is dependent on the length of the cracked ITO stripes.

In Figure 5.3 (a), when 5000 Hz 60 volts is applied on the 2 mm width copper tape, the response line width of driving image is 8 mm; in Figure 5.3 (b), when 500 Hz 60
volts is applied on the 2 mm width copper tape, the response line width of driving image is 20 mm. The line width of driving image is increased as the frequency of applied voltage is decreased. This is a very interesting phenomenon.

Similarly, as shown in Figure 5.3 (c) and (d), for the PDLC sample made by 10 cm length cracked ITO stripes, when 5000 Hz is applied, the response line width is 7 mm; when 500 Hz is applied, the response line width is 17 mm. The line width is also increased as the frequency of applied voltage is decreased.

Similarly, as shown in Figure 5.3 (e) and (f), for the PDLC sample made by 27 cm length cracked ITO stripes, when 5000 Hz is applied, the response line width is 4 mm; when 500 Hz is applied, the response line width is 13 mm. The line width is still increased as the frequency of applied voltage is decreased.
Figure 5.3: The line width of driving image is dependent on the frequency of applied voltage and the length of cracked ITO stripes. (a) (c) (e): 5000 Hz and 60 volts voltage is applied on the 2 mm adhesive copper tape. (b) (d) (f): 500 Hz and 60 volts voltage is applied on the 2 mm adhesive copper tape.
5.3 Different Lengths of Cracked ITO Stripes

In above analysis, the results are compared for the same PDLC sample in which different frequencies are applied while the length of cracked ITO stripes is identical. We can also compare the results on different PDLC samples in which identical frequency is applied while the lengths of cracked ITO stripes are different.

As shown in Figure 5.3 (a), (c) and (e), for the PDLC samples made by the cracked ITO stripes with length of 5 cm, 10 cm and 27 cm, the response line width of driving image is 8 mm, 7 mm and 4 mm respectively, while the identical frequency 5000 Hz 60 volts is applied on the 2 mm width adhesive copper tape. The line width of driving image is decreased as the length of cracked ITO stripes increased. This is also a very interesting phenomenon. Similarly, in Figure 5.3 (b), (d) and (f), the response line width is 20 mm, 17 mm and 13 mm respectively, while the identical frequency 500 Hz 60 volts is applied. The comparison of frequency dependent line widths for different lengths of cracked ITO stripes is in Table 5.1.

<table>
<thead>
<tr>
<th>Frequency (Hz)</th>
<th>Line width (mm) on PDLC of 5 cm cracked ITO stripes</th>
<th>Line width (mm) on PDLC of 10 cm cracked ITO stripes</th>
<th>Line width (mm) on PDLC of 27 cm cracked ITO stripes</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 Hz</td>
<td>35 ~ 40 (fuzzy edge)</td>
<td>32 ~ 38 (fuzzy edge)</td>
<td>25 ~ 30 (fuzzy edge)</td>
</tr>
<tr>
<td>100 Hz</td>
<td>30 ~ 35 (fuzzy edge)</td>
<td>26 ~ 32 (fuzzy edge)</td>
<td>20 ~ 25 (fuzzy edge)</td>
</tr>
<tr>
<td>500 Hz</td>
<td>20</td>
<td>17</td>
<td>13</td>
</tr>
<tr>
<td>1000 Hz</td>
<td>15</td>
<td>13</td>
<td>10</td>
</tr>
<tr>
<td>2000 Hz</td>
<td>12</td>
<td>11</td>
<td>8</td>
</tr>
<tr>
<td>3000 Hz</td>
<td>10</td>
<td>9</td>
<td>6</td>
</tr>
<tr>
<td>5000 Hz</td>
<td>8</td>
<td>7</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 5.1: The comparison of the frequency dependent line width of driving image on PDLC samples made by different lengths of cracked ITO stripes. The width of adhesive copper tape is 2 mm. The amplitude of the applied voltage is 60 volts.
There are two more things we can find in Table 5.1. The first one is the fuzzy edge of the line image observed when very low frequency, i.e. 50 Hz to 100 Hz, is applied. This fuzzy edge is not observed when higher frequencies are applied, no matter how long the length of cracked ITO stripes is. The root cause of this effect is related to the slope of voltage decay. When a low frequency is applied, the voltage decay is slow, so a fuzzy edge is observed. Similarly, when a high frequency is applied, the voltage decay is fast, so a sharp edge is observed.

The second one is the “tunability” of line width. We can define the tunability as the ratio of the maximum line width with respect to the minimum line width:

\[
\frac{W_{\text{max}}(\omega)}{W_{\text{min}}(\omega)} \equiv \text{tunability}
\]

and the “tuning ratio” as:

\[
\frac{W(\omega)}{W_{\text{min}}(\omega)} \equiv \text{tuning ratio}
\]

Table 5.2 is converted from Table 5.1, in which the tunability is equal to the tuning ratio shown in the row of 50 Hz. The tunability is increased as the length of cracked ITO stripes is increased; it means the frequency dependent behavior is more obvious for the larger size PDLC sample, which is exactly the requirement of widow application.
Table 5.2: The tuning ratio and tunability (i.e. values in the row of 50 Hz), which provide quantitative analysis for the frequency dependent line width of driving image. Three PDLC samples with different lengths of cracked ITO stripe are compared. This table is converted from Table 5.1.

<table>
<thead>
<tr>
<th></th>
<th>Tuning ratio on PDLC of 5 cm cracked ITO stripes</th>
<th>Tuning ratio on PDLC of 10 cm cracked ITO stripes</th>
<th>Tuning ratio on PDLC of 27 cm cracked ITO stripes</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 Hz</td>
<td>4.38 ~ 5</td>
<td>4.57 ~ 5.43</td>
<td>6.25 ~ 7.5</td>
</tr>
<tr>
<td>100 Hz</td>
<td>3.75 ~ 4.38</td>
<td>3.71 ~ 4.57</td>
<td>5 ~ 6.25</td>
</tr>
<tr>
<td>500 Hz</td>
<td>2.5</td>
<td>2.43</td>
<td>3.25</td>
</tr>
<tr>
<td>1000 Hz</td>
<td>1.88</td>
<td>1.86</td>
<td>2.5</td>
</tr>
<tr>
<td>2000 Hz</td>
<td>1.5</td>
<td>1.57</td>
<td>2</td>
</tr>
<tr>
<td>3000 Hz</td>
<td>1.25</td>
<td>1.29</td>
<td>1.5</td>
</tr>
<tr>
<td>5000 Hz</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>
5.4 Response Voltage at Different Positions of Cracked ITO Stripes

In section 5.2 and 5.3, the frequency dependent line width of the driving image is clearly observed. Next we will measure the amplitude and phase of response voltage at different positions in a device made by our cracked ITO on PET substrate. We want to appreciate the energetic directions and hands-on helps from Professor Yokoyama on all the processes including initial idea, measurement system, new device, and data analysis in this section.

5.4.1 Experiment Setup for Voltage Measurement

In Figure 5.4 (a) and (b), a device is made by laminating two plates, and NOA65 UV curable adhesive layer is sandwiched in between. The bottom plate is our parallel cracked ITO stripes on PET substrate, which is made by the single bending technology illustrated in chapter 2, and its resistance ratio is about 15 (i.e. 75 k ohm / 5 k ohm) measured by using a multi-meter with two probes separating 2 cm. The top plate is a continuous ITO plane on PET substrate, in which the PET substrate is facing to the NOA65 layer. The reason why we use this device rather than a PDLC window to do the response voltage measurement is because we discovered that there is still a large resistor existing in the PDLC layer of window size sample, which will make our model in chapter 6 more complicated. Our goal is to analyze the behavior of cracked ITO, therefore a simpler device and model will be a better choice.

When an external testing voltage $V \cdot \sin(\omega \cdot t)$ having amplitude $V$ (1 volt in our test) and frequency $\omega$ (125 k Hz to 1 k Hz) is applied on the 2 mm wide contact
electrode, the amplitude and phase of response voltage is measured at different positions by a probe from the measurement system of a voltage follower and a lock-in amplifier, as shown in Figure 5.4 (a) (c) and (d).

During the measurement, we discovered that the contact area of measuring probe has significant influence on the measured result. For instance, if we use 2 mm side copper tape as the probe electrode and do the measurement, because of the large value of capacitance created by this 2 mm copper tape, we measured higher value of response voltage due to this big capacitor. Our goal is to analyze the behavior of cracked ITO, and a simpler system is a better choice achieving it. So we use a sharp (around 0.5 mm) contact probe to do our measurement, as shown in Figure 5.4 (d), and there is no 2 mm wide copper tape between the testing power source and the probe.

We also discovered that a window size (40 cm x 30 cm) sample is not suitable for our measurement, because it is not completely an insulator of the PDLC layer. Several hundred of kilo ohm is measured between the cracked ITO stripe and the plan ITO layer, which means the PDLC layer is the parallel of capacitor and resistor. In order to simplify our model and analysis on cracked ITO, we made a new device as shown in Figure 5.4 (b), which is made by laminating a cracked ITO on PET substrate with a flipped plan ITO on PET substrate, and its dimension is 4 cm by 3 cm which makes the device uniform and easy to be tested.
Figure 5.4: The system and device of measuring the amplitude and phase of response voltage at different positions on the cracked ITO stripes. (a) The illustration of the measurement system including a lock-in amplifier and a voltage follower, and the top view of device. (b) The side view of device. (c) The picture of measurement system and device. (d) The picture of measuring the device by using a probe from the lock-in amplifier.

5.4.2 Measurement Result of Response Voltage

Based on the measurement system and device shown in Figure 5.4, we measured:

<table>
<thead>
<tr>
<th>Lock-in amplifier testing frequency and voltage</th>
<th>0 cm, measured voltage(%)</th>
<th>0 cm, measured phase</th>
<th>1 cm, measured voltage(%)</th>
<th>1 cm, measured phase</th>
<th>2 cm, measured voltage(%)</th>
<th>2 cm, measured phase</th>
<th>3 cm, measured voltage</th>
<th>3 cm, measured phase</th>
<th>4 cm, measured voltage(%)</th>
<th>4 cm, measured phase</th>
</tr>
</thead>
<tbody>
<tr>
<td>125 k Hz, 1 volt, self test: voltage 100.5%, phase 0</td>
<td>79.1</td>
<td>-29.8</td>
<td>78.7</td>
<td>-32</td>
<td>52.1</td>
<td>-65.3</td>
<td>31.4</td>
<td>-86.8</td>
<td>21.5</td>
<td>-118.4</td>
</tr>
<tr>
<td>90 k Hz, 1 volt, self test: voltage 101.1%, phase 0</td>
<td>91.2</td>
<td>-17.1</td>
<td>90.5</td>
<td>-19</td>
<td>63.2</td>
<td>-52.1</td>
<td>42.4</td>
<td>-74.3</td>
<td>30.5</td>
<td>-104.5</td>
</tr>
<tr>
<td>60 k Hz, 1 volt, self test: voltage 102%, phase 0</td>
<td>96.4</td>
<td>-10</td>
<td>96.2</td>
<td>-11.6</td>
<td>74.6</td>
<td>-42.4</td>
<td>57.5</td>
<td>-61.3</td>
<td>44.2</td>
<td>-86.1</td>
</tr>
<tr>
<td>30 k Hz, 1 volt, self test: voltage 103%, phase 0</td>
<td>100.6</td>
<td>-5.6</td>
<td>100.7</td>
<td>-6.3</td>
<td>92</td>
<td>-25.4</td>
<td>82.1</td>
<td>-39.7</td>
<td>73.2</td>
<td>-54.5</td>
</tr>
<tr>
<td>1 k Hz, 1 volt, self test: voltage 98.1%, phase 0</td>
<td>98.1</td>
<td>-0.2</td>
<td>98.1</td>
<td>-0.2</td>
<td>98.1</td>
<td>-0.9</td>
<td>98.1</td>
<td>-1.5</td>
<td>98.1</td>
<td>-2.2</td>
</tr>
</tbody>
</table>
Table 5.3: The amplitude and phase measurement results of response voltages at different positions of cracked ITO on PET substrate, while the frequency of applied voltage is ranging from 1 k Hz to 125 k Hz.

It is very important to calibrate and do the self-test every time before we start the measurement of each frequency. As shown in Table 5.3, after the self-test, the amplitude of testing voltage must be around 100% of 1 volt, and the phase of testing voltage must be 0. The testing voltage is applied at the position of 0 cm (point A) as shown in Figure 5.4 (d), and we can notice that the amplitude and phase has been changed from the source of lock-in amplifier to the position of 0 cm. Therefore, after the measurement, we need to divide all measured amplitudes by a factor which makes the amplitude at 0 cm to be 100%; and we also need to subtract all measured phases by a factor which makes the phase at 0 cm to be 0. After doing this modification, we can get the beautiful plots as shown in Figure 5.5 (a) to (d) which provides a set of quantitative data describing the behavior of response voltages at different positions (1 cm to 4 cm) while applying different frequencies (1 k Hz to 125 k Hz) on a device made by cracked ITO on PET substrate.

In Figure 5.5 (a), the amplitude of response voltage for different applying frequencies are plotted, when the testing position is closer to the applied voltage, the amplitude does not drop as much as the farther position at higher frequency, this is why we can see a narrower line image at higher frequency, as shown in Figure 5.5 (c).

In Figure 5.5 (b), the phase of response voltage for different applying frequencies are plotted, when the testing position is closer to the applied voltage, the phase does not
shifted as much as the farther position at higher frequency, this is an expected result caused by the equivalent RC ladder circuit we will discuss in section 6.5 and 6.6, as shown in Figure 5.5 (d).
(a) Voltage (%)

(b) Phase
Figure 5.5: The plots of amplitude and phase of response voltages at different positions of cracked ITO on PET substrate, while the frequency of applied voltage is ranging from 1 k Hz to 125 k Hz. These plots are transferred from Table 5.3 by dividing a factor to make the amplitude at 0 cm to be 100%, and subtracting a factor to make the phase at 0 cm to be 0.
CHAPTER 6

Analysis and Simulation

In order to explain the experimental results observed in CHAPTER 5, the device modeling and quantitative simulation for PDLC window made by parallel cracked ITO on PET substrate are developed in this chapter. The analysis of device modeling and the comparison between experimental and simulation results are performed, therefore the cause and effect relationship is provided to explain the behavior of frequency dependent line width of driving image in PDLC window made by parallel cracked ITO.

6.1 Analysis of RC Circuit

Before doing the simulation for frequency dependent line image and phase shift observed in cracked ITO PDLC window, we have to know the behavior of RC circuits [89–90].

The circuit of two serially connected resistances driven by an AC (i.e. alternating current) voltage is shown in Figure 6.1.

![Diagram of two serially connected resistors driven by AC voltage](image)

Figure 6.1: Two serially connected resistors driven by an AC voltage.
From Ohm’s law and Kirchhoff’s law, we have: \[ I(t) = \frac{V \cdot \sin(\omega \cdot t)}{R_1 + R_2}, \]

\[ V_1(t) = \frac{V \cdot \sin(\omega \cdot t)}{R_1 + R_2} \cdot R_1, \]

the amplitude is smaller than \( V \), but the phase \((\omega \cdot t)\) is not changed.

\[ V_2(t) = \frac{V \cdot \sin(\omega \cdot t)}{R_1 + R_2} \cdot R_2, \]

the amplitude is smaller than \( V \), but the phase \((\omega \cdot t)\) is not changed.

For the circuit of two serially connected resistances driven by an AC voltage, the amplitude of response voltage across the resistance is decreased, but the phase of response voltage is identical to the applied voltage.

The circuit of two serial capacitances driven by an AC voltage is shown in Figure 6.2.

![Figure 6.2: Two serially connected capacitors driven by an AC voltage.](image)
From the definition of capacitor and Kirchhoff’s law, we have:

\[ Q(t) = \frac{C_1 \cdot C_2}{C_1 + C_2} \cdot V \cdot \sin(\omega \cdot t) , \text{ and} \]

\[ V_1(t) = \frac{V \cdot \sin(\omega \cdot t)}{C_1 + C_2} \cdot C_2, \text{ the amplitude is smaller than } V, \text{ but the phase } (\omega \cdot t) \text{ is not changed.} \]

\[ V_2(t) = \frac{V \cdot \sin(\omega \cdot t)}{C_1 + C_2} \cdot C_1, \text{ the amplitude is smaller than } V, \text{ but the phase } (\omega \cdot t) \text{ is not changed.} \]

For the circuit consisted of two serial capacitances driven by an AC voltage, the amplitude of the response voltage across the capacitance is decreased, but the phase of the response voltage is identical to the applied voltage.

The circuit consisted of a resistance serially connected with a capacitance driven by an AC voltage is shown in Figure 6.3.

![Figure 6.3: A resistor serially connected with a capacitor driven by an AC voltage.](image)

From Ohm’s law, the definition of capacitor, and Kirchhoff’s law, we have:
\[
\frac{d}{dt} I(t) + \frac{I(t)}{R \cdot C} = \frac{V \cdot \omega \cdot \cos(\omega \cdot t)}{R} \quad \text{…. Equation 6.1}
\]

There are standard formulas of solving the first order differential equation:

For \( \frac{dy(x)}{dx} + p(x) \cdot y(x) = q(x) \), an integrating factor \( e^{\int p(x) \, dx} \) can be used to get

\[
y(x) = \left( e^{-\int p(x) \, dx} \right) \left[ \int e^{\int p(x) \, dx} \cdot q(x) \, dx + C \right] \quad \text{…. Formula 6.1}
\]

But we find that it is not easy to solve Equation 6.1 by directly applying Formula 6.1, so we solve it by guessing the format of its solution. We find that this approach is easier than applying the formula and it helps us to derive each step to get the solution.

In Equation 6.1, the right hand side is a cosine function, the left hand side is a summation of function \( I(t) \) and the derivative of itself \( \frac{d}{dt} I(t) \). The derivative of a sine function is a cosine function. The difference between a cosine function and a sine function is phase shift \( \frac{\pi}{2} \). So we guess: the format of \( I(t) \) should be a sine function, in which the phase is shifted to a specific value, but the frequency should be maintained at \( \omega \), and therefore to make the summation of \( I(t) \) and \( \frac{d}{dt} I(t) \) to be a cosine function.

Meanwhile, the amplitude of \( I(t) \) should be adjusted to fit the coefficients in Equation 6.1. So we guess the format of \( I(t) \) is: \( I(t) = I \cdot \sin\phi \cdot \sin(t + \phi) \), which is put back into Equation 6.1, and then with the help of formulas:

\[
\cos(A + B) = \cos A \cdot \cos B - \sin A \cdot \sin B
\]
\[
\sin(A + B) = \sin A \cdot \cos B + \cos A \cdot \sin B
\]

We can get:

\[
I \cdot \omega \cdot \cos \phi + \frac{I \cdot \sin \phi}{R \cdot C} = \frac{V \cdot \omega}{R} \quad \text{.... Equation 6.2}
\]

\[
I \cdot \omega \cdot (-1) \cdot \sin \phi + \frac{I \cdot \cos \phi}{R \cdot C} = 0 \quad \text{.... Equation 6.3}
\]

From Equation 6.3, we can get: \[
\cos \phi = \frac{R \cdot C \cdot \omega}{\sqrt{(R \cdot C \cdot \omega)^2 + 1}}
\]

This result can be put back into Equation 6.2, and then we can get: \[
I = \frac{V \cdot C \cdot \omega}{\sqrt{(R \cdot C \cdot \omega)^2 + 1}}
\]

Finally, the beautiful solution of \( I(t) \) is:

\[
I(t) = I \cdot \sin(\omega \cdot t + \phi) = \frac{V \cdot C \cdot \omega}{\sqrt{(R \cdot C \cdot \omega)^2 + 1}} \cdot \sin(\omega \cdot t + \tan^{-1}\left(\frac{1}{R \cdot C \cdot \omega}\right)) \quad \text{.... Solution 6.1}
\]

Solution 6.1 can be examined by using dimension analysis: the dimension of \( V \cdot C \cdot \omega \) is (voltage)-(charge/voltage)-(1/time) = (charge/time) which is current. The frequency \( \omega \) dependent behavior of the amplitude of \( I(t) \) can be illustrated by setting:

\[
\omega = \left(\frac{1}{R \cdot C}\right) \cdot x, \text{ in which } x \text{ is a dimensionless value representing } \omega,
\]

\[
\therefore \quad I = \frac{V \cdot C \cdot \omega}{\sqrt{(R \cdot C \cdot \omega)^2 + 1}} = \frac{V}{R} \cdot \frac{x}{\sqrt{x^2 + 1}}, \text{ which can be written and plotted as:}
\]
\[ I = \frac{\omega}{\left(1 + \frac{1}{R \cdot C} \right)^2} \] \[ (V/R) + 1 \] .... Solution 6.2

Figure 6.4: The quantitative relationship between \( \frac{I}{V/R} \) and \( \frac{\omega}{1/R \cdot C} \) in the circuit of a resistor \( R \) serially connected with a capacitor \( C \). \( I \) is the amplitude of response current, \( V \) is the amplitude of applying AC voltage, \( \omega \) is the frequency of applying AC voltage.

At very low frequency, i.e. \( \omega \ll \left(\frac{1}{R \cdot C}\right) \), the amplitude \( I \) of response current \( I(t) \) is very low, i.e. \( I \ll \left(\frac{V}{R}\right) \), therefore the response voltage will be mainly across the capacitor, not the resistor, so it is a low-pass filter [91] for the response voltage across capacitor in the RC circuit shown in Figure 6.3.
At very high frequency, i.e. $\omega >> \left(\frac{1}{R \cdot C}\right)$, the amplitude $I$ of response current $I(t)$ is very high, i.e. $I \approx \left(\frac{V}{R}\right)$, therefore the response voltage will be mainly across the resistor, not the capacitor, so it is a high-pass filter [92] for the response voltage across resistor in the RC circuit shown in Figure 6.3.

Therefore, the response voltages $V1(t)$ and $V2(t)$ responding to the applied voltage $V(t) = V \cdot \sin(\omega \cdot t)$ in Figure 6.3 are:

$$V1(t) = I(t) \cdot R = V \cdot \frac{x}{\sqrt{x^2 + 1}} \cdot \sin(\omega \cdot t + \tan^{-1}\left(\frac{1}{x}\right)),$$

if $\frac{\omega}{\left(\frac{1}{R \cdot C}\right)} \equiv x$ which is defined to represent the scale of $\omega$, the amplitude is smaller than $V$, and the phase $(\omega \cdot t)$ is shifted to be $(\omega \cdot t + \tan^{-1}\left(\frac{1}{x}\right))$.

$$V2(t) = V(t) - V1(t) = V \left[\sin(\omega \cdot t) - \frac{x}{\sqrt{x^2 + 1}} \cdot \sin(\omega \cdot t + \tan^{-1}\left(\frac{1}{x}\right))\right],$$

the amplitude is smaller than $V$, and the phase $(\omega \cdot t)$ is changed if $x$ is not zero, which means as long as the applied voltage is not a DC voltage, i.e. it is an AC voltage, the amplitude of $V2(t)$ will be smaller than $V$, and the phase $(\omega \cdot t)$ will be shifted.

Based on the foregoing analysis, we know that for a circuit consisting of a resistor serially connected with a capacitance, when an AC voltage is applied on, the amplitude of the response voltage across the resistor or the capacitor is decreased and the amplitude is
controlled by the frequency of applied AC voltage, and the phase of the response voltage is also shifted. These results can be used to explain the experimental results observed in CHAPTER 5.

In CHAPTER 5, the line width of driving image in PDLC made by parallel cracked ITO is controlled by the frequency of applied voltage. If we consider the simplest case, it is the circuit shown in Figure 6.3, a resistor serially connected with a capacitor, where the resistance is from parallel cracked ITO stripes used in the top or bottom plate, and the capacitance is from PDLC. At very low frequency, the response voltage across the capacitor PDLC is high, so LC is aligned by the response voltage, so the line image is observed; at very high frequency, the response voltage across the capacitor PDLC is low, so LC is not aligned by the response voltage, so the line image is not observed. **In the circuit of a resistor serially connected with a capacitor, the resistor prefers to receive the voltage from high frequency applying voltage, and the capacitor prefers to receive the voltage from low frequency applying voltage.** This is the basic physics for the frequency dependent image observed in our PDLC window, and it is very important for this thesis.

For an AC applied voltage, the amplitude of response voltage is decreased and the phase of response voltage is shifted, if there are serially connected resistors and capacitors in the circuit; but there is no phase-shift between applied voltage and response voltage in a pure resistors or pure capacitors circuit. **In CHAPTER 5, we observed the**
decreased amplitude and phase-shift of response voltage, so there should be resistors and capacitors serially connected in the model of our PDLC window.

In our PDLC window made by cracked ITO on PET substrate, the resistors are from cracked ITO stripes and tiny connections between neighboring cracked ITO stripes, the capacitances are from PDLC. Because PDLC capacitors are parallel connected in our device, if there is no resistance between them, there will be no voltage drop among these capacitors, and all PDLC capacitors will be turned on simultaneously when the AC voltage is applied.

The explanation of frequency dependent behavior of driving image is as follows. By applying an AC voltage of frequency $\omega$, the impedance [93] of a resistor having resistance $R$ is $R$, and the impedance of a capacitor having capacitance $C$ is $\frac{1}{j \cdot \omega \cdot C}$, where $j = \sqrt{-1}$. There are serially connected resistors and capacitors in our PDLC window, when the frequency $\omega$ is increased, the impedance of circuit is decreased because of the existence of capacitors, so the current is increased, so the voltage drop across resistors is increased, so the voltage is decreased more rapidly along the direction perpendicular to cracks, so the line width of driving image is decreased, because PDLC need a minimum voltage (i.e. 25 volts in our PDLC device) to be turned on. The “Cause and Effect” relationship explaining the frequency dependent line width of driving image is shown in Figure 6.5.
Figure 6.5: The “Cause and Effect” relationship explaining the frequency
dependent line width of driving image in the PDLC window made by parallel
cracked ITO on PET substrate.

In our PDLC window, the width of cracked ITO stripes is 10 microns and the line
width of driving image is several millimeters to centimeter, so at least the number of ITO
stripes need to be \( \frac{1 \cdot 10^{-2}}{10 \cdot 10^{-6}} = 10^3 \) in the circuit model, if we want to simulate the line
width of driving image. Because each one of these \( 10^3 \) ITO stripes is also corresponding
to a capacitor, therefore at least \( 2 \cdot 10^3 \) components are required in the circuit model.
6.2 Device Modeling

In order to do the simulation of our cracked ITO PDLC window, a device model is built up, and then all the values of resistance and capacitance in that model are measured, finally the software LTspice [94] is used to do the simulation. Based on the analysis in section 6.1, we know that there are many serially connected resistors and capacitors in the device model of our cracked ITO PDLC window, a specific layout of these resistors and capacitors is figured out as follows.

In Figure 6.6, the structure of PDLC window made by our parallel cracked ITO stripes on PET substrate is illustrated. During the assembling process, as shown in (a), the cracked ITO substrate is flipped over to face the plane ITO plate, the adhesive copper tape is connected with ITO at the edge on both substrates, the length of cracked ITO stripes is 10 cm, and the width of copper tape is 2 mm. The side view along the Y direction, as shown in (b), the width of cracks is 0.1 micron, which is an average as shown in Figure 2.9 and 2.14, the width of cracked ITO stripes is 10 microns, which is an average as shown in Figure 2.9 and 2.16, and the thickness of PDLC layer is 15 microns. The tiny connections between neighboring parallel cracked ITO stripes are illustrated as shown in (c). We want to report that the tiny and uniform connections shown in Figure 6.6 (b) and (c) are representative symbols of the basically unknown structure by which two neighboring cracked ITO stripes are connected. We don’t have pictures to illustrate the real structure of tiny connections. One possibility of the structure is a fully cracked
ITO which is partially contacted at the bottom, because we know our ITO are fully cracked on PET substrate according to the Beuth’s model introduced in Chapter 4.

In order to simplify the unknown structure into an analyzable model, we assume that the resistance is isotropic (i.e. identical along all directions) in the tiny connections area (i.e. the crack between two neighboring ITO stripes) in our model. We appreciate Professor Yokoyama’s directions on the assumptions of our model, by which we are allowed to explain the model more clearly.
Figure 6.6: The structure of PDLC window made by our parallel cracked ITO stripes on PET substrate. (a) In the assembling process, the cracked ITO substrate is flipped over to face the plane ITO substrate. (b) The side view of PDLC window along the Y direction, the dimensions of cracks, cracked ITO stripes, and each layer are illustrated. (c) The layout of adhesive copper tape on top substrate and the tiny connections between neighboring parallel cracked ITO stripes. The tiny and uniform connections shown in Figure 6.6 (b) and (c) are representative symbols of the basically unknown structure by which two neighboring cracked ITO stripes are connected.
The device modeling (i.e. equivalent circuit) of cracked ITO PDLC window is figured out as shown in Figure 6.7. The resistors of cracked ITO stripes are labeled as 1, 2, 3, and so on, the resistors of the tiny connections between two neighboring cracked ITO stripes are labeled as A, B, C, and so on. We want to report that this device modeling is a very simple one which only consider the capacitance of PDLC between cracked ITO stripes and the plane ITO layer, the capacitances between neighboring ITO stripes are not considered, but they are very important and necessary if we want to get more accurate and advanced simulation results. This is the insufficient part of our model. We appreciate Professor Yokoyama’s directions on the capacitances between neighboring ITO stripes, which enlighten the insufficient part of our model, and provide an approach to upgrade it to a higher level of accuracy.
Figure 6.7: The device modeling (i.e. equivalent circuit) of PDLC window made by cracked ITO on PET substrate.

The reason why this equivalent circuit is a modeling for our cracked ITO PDLC device is explained as follows. The current flows from applied voltage source through the copper tape into the central cracked ITO stripes 1 along the direction parallel to cracks. The current flow direction is important, because it determines the value of resistance in the equivalent circuit. Then the current will reach the central capacitor C1 which is defined by an ideal capacitor which is composed of two ideal electrodes (i.e. resistance is zero) which have an identical area size of central cracked ITO stripes 1, and
the PDLC of 15 microns thickness is sandwiched between these two ideal electrodes. It is important to have the idea of ideal capacitor in the equivalent circuit; because it simplifies the PDLC window from a complicated device into a simple model consisted of resistors and capacitors. Then the displacement current flows into the plane ITO on the bottom substrate. We assume the resistance of plane ITO is close to zero on the bottom substrate.

To determine the position of resistor representing the slight ITO connections between cracks is important for building up the equivalent circuit. In our model, after the current flowing through the central cracked ITO stripes 1, there are two possible ways in front of it: one is to flow into the first crack A, and the other one is to flow into the first capacitor C1, as shown in Figure 6.7.

Then the current flows into the second resistance 2, but this time the current flow direction is perpendicular to the cracks, which means the resistance value of the second resistor 2 will be very small, comparing with the first resistor 1. It is important to have a careful consideration of practical current flow directions in the device and use it to build up a reasonable modeling.

Then the current flows into the resistor B and capacitor C2, this process is similar to the condition of the first crack A and the first capacitor C1. The following steps just repeat the abovementioned steps, so an equivalent circuit is successfully constructed as shown in Figure 6.7.
6.3 Measurement of Sheet Resistance

After constructing the device modeling in Figure 6.7, the resistance values of resistor 1, 2, 3 and resistor A, B, C are required for the next step of simulation. For resistor 1, 2, 3, they are long stripes (10 cm × 10 µm) of cracked ITO, their resistance values can be calculated if the sheet resistance is known. We know that the sheet resistance of plane ITO film on PET substrate is 60 ohm per square, which is provided by the manufacturing company. But the sheet resistance of our cracked ITO long stripes should be larger than 60 ohm per square, because our ITO cracking process is not perfect. For instance, in Figure 2.9, the intersection of two neighboring cracks will appear if they are not perfectly created (i.e. not completely parallel) during the ITO cracking process. Similarly, a slightly non-parallel crack will occur, for instance, the third crack counting from left hand side or the second crack counting from right hand side in Figure 2.9. All these imperfect (i.e. not precisely parallel) cracks will increase the sheet resistance value of our cracked ITO stripes.

Four-point probe [111] is a method of measuring sheet resistance, it is operated by using two point probes of a constant current source and two point probes of voltmeter to avoid the contact resistance. The reason we didn't use it is because the sample need to be a uniform and isotropic film for this method, but our cracked ITO film is anisotropic film. During the survey of four-point probe method, we studied the van der Pauw method [112], which gave us the idea of cutting samples into squares and measuring them directly.
The plane ITO film on PET substrate is bought from Sheldahl (Accentia, 5 mil thickness of PET), and the sheet resistance of the ITO film is 60 ohm per square. By using the line contact electrodes shown in Figure 6.8 (a) and square resistors shown in Figure 6.8 (b) and (c), the sheet resistance 60 ohm per square of plane ITO film on PET substrate is correctly measured.
What we have done during the measurement of using the line contact electrode is described as follows. When we measure the resistance, initially the measured value is higher and unstable, because we did not completely press the line contact electrodes on the sample surface; after we press them on the surface tightly, the resistance will be lowered to a stable value. We do the measurement of each value for 5 to 10 times, and observe if the measured values are reproducible in the measured range; according to our observation, the error of our measured resistance is in the range of 10~30 ohm per square. The limitation of our measurement is that the accuracy of measured resistance did not reach the level of 1 ohm per square. An advanced method of measuring the accurate sheet resistance should be applying a secured contact between the line contact electrodes and the surface of our cracked ITO on PET substrate, and the possible methods include using silver paste, conductive rubber, inkjet printing, and vacuum deposition, etc. We
appreciate Professor Yokoyama’s directions on this issue of secured contact and the solutions of making these secured contacts.

The sheet resistances of parallel cracked ITO stripes on PET substrate are measured by cutting the samples into square of 1 cm × 1 cm, 2 cm × 2 cm and 3 cm × 3 cm, and using the line contact electrodes of 1 cm, 2 cm and 3 cm, respectively.

In order to identify the resistance parallel and perpendicular to the direction of cracks, two specific sheet resistances $R_{\text{para}}$ and $R_{\text{perp}}$ are defined as shown in Figure 6.9, the value of $R_{\text{para}}$ should be much smaller than $R_{\text{perp}}$, because the resistance of ITO stripes is much smaller than that of cracks.

Figure 6.9: (a) The dimensions of cracked ITO stripes and cracks. (b) The definition of sheet resistance $R_{\text{para}}$ and $R_{\text{perp}}$, where $R_{\text{para}}$ is parallel and $R_{\text{perp}}$ is perpendicular to the direction of ITO strips.
The measured sheet resistances are shown in the Table 6.1. In order to get more information of the sheet resistance, when the separation between line contact electrodes is 1 cm, the resistances are also measured.

<table>
<thead>
<tr>
<th>Square of cracked ITO on PET substrate (cm×cm)</th>
<th>1×1</th>
<th>2×2</th>
<th>2×2</th>
<th>3×3</th>
<th>3×3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length of line contact electrodes (cm)</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Separation between line contact electrodes (cm)</td>
<td>1</td>
<td>2</td>
<td>1</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>$R_{\text{para}}$ (ohm)</td>
<td>130</td>
<td>160</td>
<td>75</td>
<td>180</td>
<td>55</td>
</tr>
<tr>
<td>$R_{\text{perp}}$ (ohm)</td>
<td>1200k</td>
<td>1600k</td>
<td>750k</td>
<td>1800k</td>
<td>500k</td>
</tr>
</tbody>
</table>

Table 6.1: The measured sheet resistances, $R_{\text{para}}$ and $R_{\text{perp}}$, by using different lengths of line contact electrodes and different sizes of cracked ITO squares.

In Table 6.1, we find that the measured sheet resistance of 1 cm ×1 cm cracked ITO is 130 ohm per square, which is more than twice the value of intrinsic ITO sheet resistance 60 ohm per square. This is due to the imperfection of our cracking ITO process discussed in page 121.

Based on the measured values in Table 6.1, the values of sheet resistance of cracked ITO on PET substrate are calculated as follows. Four resistances, $R_{\text{ITO,para}}$, $R_{\text{ITO,perp}}$, $R_{\text{crack,para}}$, and $R_{\text{crack,perp}}$, are defined as illustrated in Figure 6.10 (a) to (d). The red region is ITO stripe, and the white area between two neighboring ITO stripes is crack stripe. The values of four resistances are determined by the dimension of ITO or crack stripe and the current direction indicated by green arrow. In Figure 6.14 (e), the ITO or
crack stripe is viewed as an integral of squares and therefore the sheet resistance of ITO or crack can be used to calculate these four resistances.
Figure 6.10: The definition of $R_{ITO,\text{para}}$ (a), $R_{ITO,\text{perp}}$ (b), $R_{\text{crack,para}}$ (c), $R_{\text{crack,perp}}$ (d), and how to calculate their values by using the measured sheet resistance of ITO or crack (e).

For 1 cm length and 10 $\mu$m width ITO stripe, it can be viewed as a row of 10 $\mu$m $\times$ 10 $\mu$m squares, and the sheet resistance of intrinsic non-cracked ITO is 60 ohm per square. From the Ohm’s law and the defined current direction, ideally:

$$R_{ITO,\text{para}} = 60 \Omega \cdot \frac{1\text{cm}}{10\mu\text{m}} = 6 \cdot 10^4 \Omega,$$ for 1 cm length intrinsic non-cracked ITO.

$$R_{ITO,\text{perp}} = 60 \Omega \div \frac{1\text{cm}}{10\mu\text{m}} = 6 \cdot 10^{-2} \Omega,$$ for 1 cm length intrinsic non-cracked ITO.

These two ideal values can be checked with the measured ones in Table 6.1, in which $R_{\text{para}}$ is measured as 130 $\Omega$ for 1 cm $\times$ 1 cm cracked ITO.

So: $(R_{\text{para}} = 130 \Omega) \equiv (R_{ITO,\text{para}} \div \frac{1\text{cm}}{10\mu\text{m}})$

$$\Rightarrow R_{ITO,\text{para}} \approx 13 \cdot 10^4 \Omega,$$ which is $\frac{13}{6} \approx 2.17$ times larger than the intrinsic non-cracked value of $6 \cdot 10^4 \Omega$. This is due to the imperfection of our cracking ITO process discussed
in page 121. In other words, for the sheet resistance of ITO, \( R_{\text{sheet,ITO}} \), it is 60 ohm per square on the intrinsic non-cracked ITO, and it is increased to 130 ohm per square on our cracked ITO.

For 1 cm length and 0.1 \( \mu \text{m} \) width ITO stripe, it can be viewed as a row of 0.1 \( \mu \text{m} \times 0.1 \mu \text{m} \) squares, and the sheet resistance of each square needs to be calculated. If the sheet resistance of crack is defined as \( R_{\text{sheet,crack}} \), from the Ohm’s law and the defined current direction, ideally:

\[
R_{\text{crack,para}} = R_{\text{sheet,crack}} \cdot \frac{1 \text{cm}}{0.1 \mu \text{m}} = R_{\text{sheet,crack}} \cdot 10^5, \quad \text{for 1 cm length crack.}
\]

\[
R_{\text{crack,perp}} = R_{\text{sheet,crack}} \cdot \frac{1 \text{cm}}{0.1 \mu \text{m}} = R_{\text{sheet,crack}} \cdot 10^{-5}, \quad \text{for 1 cm length crack.}
\]

These two ideal values can be checked with the measured ones in Table 6.1, in which \( R_{\text{perp}} \) is measured as 1200k for 1 cm \( \times \) 1 cm cracked ITO.

So: \( (R_{\text{perp}} = 1200k\Omega) = \left( R_{\text{ITO,perp}} \cdot \frac{1 \text{cm}}{10 \mu \text{m}} + R_{\text{crack,perp}} \cdot \frac{1 \text{cm}}{10 \mu \text{m}} \right) \) where \( R_{\text{ITO,perp}} = 13 \cdot 10^{-2} \Omega \)

\Rightarrow R_{\text{crack,perp}} = 1200\Omega, \quad \text{for 1 cm length crack.}

So: \( R_{\text{crack,perp}} = R_{\text{sheet,crack}} \cdot 10^{-5} \) where \( R_{\text{crack,perp}} = 1200\Omega \)

\Rightarrow R_{\text{sheet,crack}} = 1.2 \cdot 10^8 \Omega

So: \( R_{\text{crack,para}} = R_{\text{sheet,crack}} \cdot 10^5 = 1.2 \cdot 10^{13} \Omega \), which is an open circuit, for 1 cm length crack.
In summary, from the measured values in Table 6.1 and foregoing calculations, for the cracked ITO on PET substrate:

1. \( R_{\text{sheet,ITO}} = 120 \Omega \sim 180 \Omega \), the average is 150\( \Omega \).

2. \( R_{\text{sheet,crack}} = 1.2 \cdot 10^8 \Omega \sim 1.8 \cdot 10^8 \Omega \), the average is 1.5\( \cdot 10^8 \Omega \).

3. \( \frac{R_{\text{sheet,crack}}}{R_{\text{sheet,ITO}}} = 10^6 \)

4. For length and 10 \( \mu m \) width ITO stripe:

\[
\begin{align*}
R_{\text{ITO, para}} &= R_{\text{sheet,ITO}} \cdot \frac{L}{10 \mu m} \\
R_{\text{ITO, perp}} &= R_{\text{sheet,ITO}} \div \frac{L}{10 \mu m} \ldots \text{ Equation 6.4}
\end{align*}
\]

5. For length and 0.1 \( \mu m \) width crack stripe:

\[
\begin{align*}
R_{\text{crack, para}} &= R_{\text{sheet,crack}} \cdot \frac{L}{0.1 \mu m} \cong \text{open circuit} \\
R_{\text{crack, perp}} &= R_{\text{sheet,crack}} \div \frac{L}{0.1 \mu m} \ldots \text{ Equation 6.5}
\end{align*}
\]

If we want to use an alternative patterning technology (e.g. photo-lithography or laser patterning) to make slightly connected ITO stripes on PET substrate which is identical to the cracked ITO made by our cracking processes, the required dimension of narrow ITO connection between ITO stripes is determined by Equation 6.4 and Equation 6.5. The length of ITO stripes or cracks is \( L \), the width of ITO stripes is 10 \( \mu m \), and the width of cracks is 0.1 \( \mu m \). Assuming the width of a narrow ITO connection which has
the length of 0.1 µm between two neighboring ITO stripes is \( x \), no matter how long \( L \) is, we have:

\[
\frac{R_{\text{crack,perp}}}{R_{\text{ITO,perp}}} = \frac{R_{\text{sheet,crack}}}{R_{\text{sheet,ITO}}} = \frac{0.1 \mu m}{10 \mu m} = 10^6 \cdot 10^{-2} = 10^4
\]

If \( L = 10 \text{ cm} \), \( \Rightarrow \frac{(0.1 \mu m / x)}{(10 \mu m / 10 \text{ cm})} = 10^4 \Rightarrow x = 0.1 \mu m \)

If \( L = 100 \text{ cm} \), \( \Rightarrow \frac{(0.1 \mu m / x)}{(10 \mu m / 100 \text{ cm})} = 10^4 \Rightarrow x = 1 \mu m \)

For the wafer size application (i.e. \( L = 10 \text{ cm} \)), the challenge of photo-lithography process is 0.1 µm, which is the width of cracks or the width of narrow connection. In this case, our cracking ITO process is more economic comparing with photo-lithography process, because the cost of 0.1 µm size mask and chemical wet etching process are saved.

For the window size application (i.e. \( L = 100 \text{ cm} \)), the challenge of laser patterning process is 0.1 µm, which is the width of cracks, because the width of the narrow connection is 1µm. In this case, our cracking ITO process is a unique solution, because 0.1 µm is not achievable by currently commercialized laser patterning process in the window size (i.e. 100 cm) application. The resolution of ITO pattern and the field area achievable by currently commercialized laser patterning process are as follows: for the dry ablating ITO, a field area of 0.25 m × 0.25 m, on which the laser spot dimension is down to 10 µm, or a field area of 1.6 m × 1.6 m on which the laser spot dimension is increased to approximately 100 µm [95–96].
6.4 Measurement of Capacitance

In Table 6.2, the capacitance of PDLC made in our cracked ITO windows is measured by using a digital capacitance meter CHY15 at the testing frequency of 820 Hz.

In Figure 6.11, the capacitance of a capacitor is defined as:

\[
C \equiv \frac{Q}{V} = \varepsilon_r \cdot \varepsilon_0 \cdot \frac{\text{Area}}{\text{Distance}} = \varepsilon_r \cdot \varepsilon_0 \cdot \frac{L \cdot W}{d},
\]

\[
\varepsilon_0 = 8.854 \times 10^{-12} \text{ F} \cdot \text{m}^{-1},
\]

which is the electric constant or the vacuum permittivity.

\[
\varepsilon_r = 1 ,
\]

vacuum is in-between two ideal conductive plates.

In our model, for the PDLC corresponding to a cracked ITO stripe, the volume is 10 cm × 10 μm × 15 μm, and the capacitance is:

\[
\frac{10 \cdot 10^{-6}}{9 \cdot 12.5 \cdot 10^{-5}} \cdot 69.5 \text{nF} = 6.17 \text{pF}
\]

\[
\frac{10 \cdot 10^{-6}}{5 \cdot 5 \cdot 10^{-2}} \cdot (15.8 \sim 16.2) \text{nF} = (6.32 \sim 6.48) \text{pF}
\]

So 6.3 pF is used as the capacitance of PDLC which has the volume of 10 cm × 10 μm × 15 μm in our model.

<table>
<thead>
<tr>
<th>Volume of PDLC samples: Length (cm) × Width (cm) × Distance (μm)</th>
<th>Measured Capacitance (nF) of PDLC samples</th>
<th>Calculated Capacitance (pF) for PDLC having volume of 10 cm × 10 μm × 15 μm, which is responding to a cracked ITO stripes in our model</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.5 × 9 × 15</td>
<td>69.5</td>
<td>6.17</td>
</tr>
<tr>
<td>5 × 5 × 15</td>
<td>15.8 ~ 16.2</td>
<td>6.32 ~ 6.48</td>
</tr>
</tbody>
</table>

Table 6.2: The capacitance measurement of PDLC samples, and the calculation of PDLC capacitance corresponding to a cracked ITO stripe in our model.
Figure 6.11: The value of capacitance is determined by the structure and dimensions of a capacitor.

6.5 LTspice Simulation

In Figure 6.12, the equivalent circuit of PDLC window made by cracked ITO on PET substrate is illustrated, and the length of cracks is 10 cm in this model.

The value of resistance 1 is calculated as follows. The width of adhesive copper tape is 2 mm, and the current flow is along 10 cm ITO stripes, so:

\[
R_{ITO,para} \approx R_{sheet,ITO} \frac{10\text{cm}}{10\mu\text{m}} + \frac{2\text{mm}}{10\mu\text{m}} = 7.5 \cdot 10^3 \Omega, \text{ where } R_{sheet,ITO} = 150 \Omega
\]

The value of capacitance 1 is calculated as follows. The width of adhesive copper tape is 2 mm, and the length of ITO stripes is 10 cm, so the capacitance of PDLC having volume of 10 cm $\times$ 2 mm $\times$ 15 $\mu$m is:

\[
6.3\text{pF} \cdot \frac{2\text{mm}}{10\mu\text{m}} = 1260\text{pF}
\]

The value of resistance A or B or C is calculated as follows. The width of the crack is 10 cm, and the current flow is along 0.1 $\mu$m crack, so:

\[
R_{crack,perp} = R_{sheet,crack} \frac{10\text{cm}}{0.1\mu\text{m}} = 150\Omega, \text{ where } R_{sheet,crack} = 1.5 \cdot 10^8\Omega
\]
The value of resistance 2 or 3 or 4 is calculated as follows. The width of the ITO is 10 cm, and the current flow is along 10 μm ITO, so:

\[ R_{ITO,\,perp} = R_{sheet,\,ITO} \times \frac{10\,cm}{10\,\mu m} = 0.015\,\Omega, \text{ where } R_{sheet,\,ITO} = 150\,\Omega \]

![Diagram of the equivalent circuit](image)

**Figure 6.12:** The equivalent circuit of PDLC window made by cracked ITO on PET substrate, in which the length of cracks is 10 cm.

In the equivalent circuit, the central line is resistor 1 and capacitor C1, and the left hand side and the right hand side are symmetric and parallel-connected, so one of them is removed to simplify the equivalent circuit. Meanwhile, 0.015 ohm is too small comparing with 150 ohm when they are serial-connected, so all resistors of 0.015 ohm
are removed. A model consisting of 200 resistors of 150 ohm and 200 capacitors of 6.3 pF is calculated by using LTspice software, as shown in Figure 6.13.
Figure 6.13: (a) The equivalent circuit consisting of 200 resistors of 150 ohm and 200 capacitors of 6.3 pF. (b) An enlarged picture for the circuit in red line region of (a). (c) The simulation result of this equivalent circuit by using LTspice: the green line is the applied voltage 60 volts 5000 Hz sine wave, the dark blue line is the response voltage of the capacitor of 1260 pF, the red line is the response voltage of the 100th capacitor of 6.3 pF, and the light blue line is the response voltage of the 200th capacitor of 6.3 pF.

Although there are only 200 resistors and 200 capacitors, the identical frequency, the voltage drop and the phase shift of response voltages are clearly observed, which is consistent with the analysis result discussed in section 6.1.

A model consisting of 1000 resistors of 150 ohm and 1000 capacitors of 6.3 pF is calculated by using LTspice software, as shown in Figure 6.14.
Figure 6.14: The equivalent circuit consisting of 1000 resistors of 150 ohm and 1000 capacitors of 6.3 pF. The green line is the applied voltage 60 volts 5000 Hz sine wave, the dark blue line is the response voltage of the capacitor of 1260 pF, the red line is the response voltage of the 200th capacitor of 6.3 pF, the light blue line is the response voltage of the 400th capacitor of 6.3 pF, the pink line is the response voltage of the 600th capacitor of 6.3 pF, the gray line is the response voltage of the 800th capacitor of 6.3 pF, and the dark green line is the response voltage of the 1000th capacitor of 6.3 pF.

For the model of 1000 resistors and 1000 capacitors, the identical frequency, the voltage drop and the phase shift of response voltages can be much more clearly observed. Besides, because the turn-on voltage of PDLC is 25 volts, so according to the simulation result shown in Figure 6.14, we can expect that the boundary of line image should be at the position of 400th capacitor of 6.3 pF, which means the width of line image should be 8 mm. By checking with Table 5.1, the observed width of line image driven by 5000 Hz 60 volts is 7 mm, which is 1 mm thinner than the simulated width 8 mm as shown in Figure
The difference between the observed and simulated results is caused by the insufficient number of resistors and capacitors in the model, which is limited by the calculation capability of our computer. By comparing the simulation results in Figure 6.13 and 6.14, we can expect that a more accurate simulation result will be obtained if the number of resistors and capacitors can be expended larger than 1000.

We can also consider the approach of “Lumped Transmission Line” [97–100] to analyze our model. Transmission lines are very important in the power transmission in today’s world, for instance, transmitting the electric power from the power station to our homes, or transporting the electric current and voltage along the circuits between ICs on the printed circuit boards. The transmission line circuit is very similar to our model of PDLC window made by cracked ITO on PET substrate as shown in Figure 6.13 (b). The circuit element (a) and symmetrical circuit (b) of lumped-element transmission line are illustrated in Figure 6.15. We can notice that Figure 6.15 (b) will be very similar to our model in Figure 6.13 (b) if the inductors $L_o$ are replaced by the resistors $R$, except that the number of components is different.
Based on the circuit structure shown in Figure 6.15, a standard wave equation [97] can be derived to represent the voltage and current behavior in the lumped-element transmission line. It means there are forward and backward traveling waves existing on the transmission line, and the voltage or current at any node is the superposition of these two waves. A critical frequency \( \omega_0 = \frac{2}{\sqrt{L_0C_0}} \), called the cutoff frequency, which determines the wave behavior in the lumped-element transmission line. If the frequency is lower than \( \omega_0 \), the wave will be lossless, i.e. the amplitude of voltages is a constant throughout the line; if the frequency is higher than \( \omega_0 \), the wave will be decayed, i.e. the amplitude will be decreased along the line, which is similar to the decayed amplitude of voltages as shown in Figure 6.14.

By considering the approach of “Lumped Transmission Line”, more possibilities are provided to the application of cracked ITO PDLC window. For instance, external inductors, capacitors, and resistor can be designed and connected to specific positions on the edge of cracked ITO PDLC window; based on the lumped-element transmission line.
approach discussed above, there should be a lossless mode available for the cracked ITO PDLC window, and it would be an improvement of saving the required energy.

6.6 Analytical Solution of Cracked ITO Device

There is an advanced approach available for solving the response voltage and current at any position of a device made by parallel cracked ITO on PET substrate: the matrix calculation. By using the Kirchhoff’s voltage and current laws and calculating several required matrixes, a clear analytical solution can be derived. We want to appreciate the energetic directions and hands-on helps from Professor Yokoyama on all the results developed in this section.

The electrical properties of the cracked ITO can be represented by the ladder array of the following equivalent circuit:

![Equivalent Circuit](image)

**Figure 6.16:** The equivalent circuit of cracked ITO PDLC device, which is a ladder array of this primary building block.

This is for the primary building block consisting of the crack and the ITO strip with the back plane ITO electrode. Here, $R_C$ and $C_C$ are the resistance and the capacitance of the crack, respectively, $C_L$ is the capacitance between the ITO strip and
the back plane electrode through the liquid crystal, and $R_e$ is the resistance of the ITO strip. The electrical characteristic of this RC circuit is described in the $2 \times 2$ matrix form:

$$
\begin{pmatrix}
V_2 \\
I_2
\end{pmatrix} = T
\begin{pmatrix}
V_1 \\
I_1
\end{pmatrix}
$$

$$
T = \begin{pmatrix}
1 & -R_e \\
0 & 1
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
-j\omega C_L & 1
\end{pmatrix}
\begin{pmatrix}
R_e \\
1 + j\omega C_R R_e
\end{pmatrix} = \begin{pmatrix}
1 + j\omega C_L R_e & -R_e - R_c \frac{1 + j\omega C_L R_e}{1 + j\omega C_R R_e} \\
-j\omega C_L & 1 + j\omega C_R R_e
\end{pmatrix}
$$

where the matrix $T$ is obtained by calculating three sub-matrixes using the Kirchhoff’s voltage and current laws on three sub-blocks in Figure 6.16.

The $n+1$ array of crack-ITO strip is described by

$$
\begin{pmatrix}
V_{n+1} \\
I_{n+1}
\end{pmatrix} = T^n
\begin{pmatrix}
V_1 \\
I_1
\end{pmatrix}
$$

$$
T^n
\begin{pmatrix}
V_{n+1} \\
I_{n+1}
\end{pmatrix} = \begin{pmatrix}
V_1 \\
I_1
\end{pmatrix}
$$

where the matrix $T^{-1}$ is the inverse of $T$ given by

$$
T^{-1} = \begin{pmatrix}
1 & \frac{R_e}{1 + j\omega C_R R_e} \\
0 & \frac{1 + j\omega C_L R_c}{1 + j\omega C_R R_e}
\end{pmatrix}
\begin{pmatrix}
1 & 0 \\
-j\omega C_L & 1
\end{pmatrix}
\begin{pmatrix}
R_e \\
1 + j\omega C_R R_e
\end{pmatrix} = \begin{pmatrix}
1 + j\omega C_L R_c & R_e + R_c \frac{1 + j\omega C_L R_c}{1 + j\omega C_R R_e} \\
-j\omega C_L & 1 + j\omega C_R R_e
\end{pmatrix}
$$

We assume that the whole system of cracked ITO consists of $N+1$ crack-strip pairs with the terminal being open.

$$
T^N
\begin{pmatrix}
V_{N+1} \\
0
\end{pmatrix} = \begin{pmatrix}
V_1 \\
I_1
\end{pmatrix}
$$
Then, the input voltage and the input current are written as

\[ V_1 = (T^N)_{11} \cdot V_{N+1} \quad \text{and} \quad I_1 = (T^N)_{21} \cdot V_{N+1} = \frac{(T^N)_{21}}{(T^N)_{11}} V_1 \]

\( \frac{(T^N)_{11}}{(T^N)_{21}} \) gives the input impedance of the system. Using these expressions, we obtain the following equation for the voltage and the current at an arbitrary strip position:

\[
\begin{pmatrix} V_{n+1} \\ I_{n+1} \end{pmatrix} = T^n \begin{pmatrix} V_1 \\ I_1 \end{pmatrix} = V_1 T^n \begin{pmatrix} 1 \\ (T^N)_{21} \end{pmatrix} = \frac{1}{(T^N)_{11}} V_1 T^n \left( T^{(N-n)} \right)_{11} \begin{pmatrix} 1 \\ 0 \end{pmatrix}
\]

It follows from this equation that

\[ V_{n+1} = \frac{(T^{(N-n)})_{11}}{(T^N)_{11}} V_1 \quad \text{and} \quad I_{n+1} = \frac{(T^{(N-n)})_{21}}{(T^N)_{11}} V_1 \]

The n-th power of the matrix \( T \) and \( T^{-1} \) can be calculated by diagonalizing the matrix as

\[ T = A \begin{pmatrix} a & 0 \\ 0 & b \end{pmatrix} A^{-1} \quad \text{and} \quad T = A \begin{pmatrix} \frac{1}{a} & 0 \\ 0 & \frac{1}{b} \end{pmatrix} A^{-1} \]

Then one obtains
The frequency roll-off behaviors of the equivalent circuit can be understood easily from the above equations without carrying out detailed calculations.
CHAPTER 7

Summary

This dissertation illustrates the development of a novel smart window technology, which establishes a revolutionary, simple, and low cost manufacturing method to make parallel cracked ITO stripes on flexible PET substrate. The flexible PET substrate coated by thin layer of ITO is processed to create parallel ITO cracks, and the resulting cracked ITO on PET is used as a substrate in polymer dispersed liquid crystal (PDLC) windows. It creates a virtual “venetian blind” effect, where the amount of light passing through the window is controlled by the frequency and amplitude of the applied voltage. This work has produced 1 journal paper [101], 4 conference papers [102 – 105], 3 US patent applications [106 – 108] and launched a start-up company, Flexible ITO Solutions (FITOS) LLC, in 2013. FITOS has been approved to get $200,000 new funding investment from State of Ohio and the GLIDE Innovation Fund in June, 2014.

7.1 Dissertation Contribution

Several contributions have been provided in this dissertation. The ideas, trials, issues, and solutions of utilizing parallel cracked ITO on PET substrates are created and implemented in CHAPTER2. PDLC and ChLCD samples made by the stretching process successfully prove the possibility of utilizing parallel cracked ITO as a solution for preparing small size electro-optics devices in CHAPTER2. The creation of multiple cracking processes successfully prove the possibility of utilizing parallel cracked ITO as
a solution for manufacturing large size electro-optics window shutter product in CHAPTER3. The root cause of cracks generated by extension stretching on substrates is studied by using solid mechanics models in CHAPTER4. The discovery of unique frequency controlled line width of driving image is a major contribution in this dissertation, as shown in CHAPTER5. A physics and electronics model of PDLC window shutter made by cracked ITO on PET substrate is well established and both analytical solution and LTspice simulation are successfully performed to explain the frequency dependent behavior observed in experiments, as shown in CHAPTER6.

7.2 Future Study Topics of Cracked ITO

(1) Circular Cracked ITO for Liquid Crystal Lens, to investigate the possibility of cracked ITO for LC lens which have significant market potential for optics industry.

(2) How to manufacture completely isolated cracked ITO on plastic film.

(3) Parallel Cracked ITO on other plastic films, to study the intrinsic reason which determines different patterns for parallel cracked ITO on different plastic films.

(4) The real pictures and profile of the slightly connected ITO structures between neighboring cracked ITO stripes, and the resistance and capacitance properties of it.

(5) More analysis on the frequency-dependent response of Cracked ITO PDLC Window. For instance, two dimensional modeling by using LTspice, and lossless design by using Lumped-Element Transmission Line approach.
(6) New application in fine angle non-mechanical beam steering system [109]. For instance, the device of liquid crystal on silicon (LCoS) optical phased array (OPA) needs one dimensional array of tens of thousands of thin electrodes (1~2 microns).

(7) New applications in commercialized products. For instance, touch screen or touch panel, especially for the large area application. Another instance is to provide partial erase function in ChLCD writing tablet, which will be very important for large area application in replacing black board or white board in the classroom or conference room.
REFERENCES


A. Kennelly, Impedance, AIEE, 1893.


[102] John L. West* and Da-Wei Lee*, “Selectively switching PDLC window made by cracked ITO on PET substrate” *SID Symposium Digest of Technical Papers*, 2014. (Note: this paper had been withdrawn by ourselves to publish a new paper)


