



國立中山大學物理學系

博士論文

Department of Physics

National Sun Yat-sen University

Doctoral Dissertation

前瞻有機與金屬氧化物

薄膜電晶體之電性分析與可靠度研究

Investigation of Reliability on Advanced Organic and Metal

Oxide Thin-film Transistors

研究生：洪揚豪

Yang-Hao Hung

指導教授：張鼎張 博士

Dr. Ting-Chang Chang

中華民國 112 年 1 月

January 2023



國立中山大學物理學系

博士論文

Department of Physics

National Sun Yat-sen University

Doctoral Dissertation

前瞻有機與金屬氧化物

薄膜電晶體之可靠度研究

Investigation of Reliability on Advanced Organic and Metal Oxide

Thin-film Transistors

研究生：洪揚豪

Yang-Hao Hung

指導教授：張鼎張 博士

Dr. Ting-Chang Chang

中華民國 112 年 1 月

January 2023

論文審定書

國立中山大學研究生學位論文審定書

本校物理學系博士班

研究生洪揚豪 (學號: D062620002) 所提論文

前瞻有機與金屬氧化物 薄膜電晶體之可靠度研究
Investigation of Reliability on Advanced Organic and Metal Oxide Thin-film Transistors

於中華民國 111 年 12 月 11 日經本委員會審查並舉行口試，符合博士學位論文標準。

學位考試委員簽章：

召集人 莫亦先 莫亦先 委員 張鼎張 張鼎張

委員 陳紀文 陳紀文 委員 蔡宗鳴 蔡宗鳴

委員 陳柏勳 陳柏勳 委員 _____

委員 _____ 委員 _____

委員 _____ 委員 _____

指導教授(張鼎張) 張鼎張 (簽名)

論文公開授權書

國立中山大學博碩士論文公開授權書



etd-1126122-170841 2022-12-26 18:08:37

本授權書所授權之論文為授權人 洪揚豪 在 國立中山大學物理學系 111 學年度 第 1 學期 取得 博士 學位之論文。

論文題目： 前瞻有機與金屬氧化物 薄膜電晶體之可變度研究

指導教授： 張鼎張

注意事項：

1. 依本校109年4月29日108學年度第2學期第6次行政會議修正通過，研究所畢業生可於上傳電子論文時，與指導教授討論後選擇學位論文紙本及電子檔之開放年限，紙本論文若選擇「四至五年後公開」者，電子論文若選擇「四至五年後公開」或「其他」者，應提供涉及機密、專利事項或依法令規定限制公開之證明資料，經指導教授及系所(學程)主管認定始能作上述選擇。
2. 因專利申請涉及論文公開時間，為避免因喪失新穎性而無法申請專利，請各位老師及同學至經濟部智慧財產局網站參考「專利各項申請案件處理時限表」後再選定論文公開時間。
另有關著作權相關資訊，請參考「經濟部智慧財產局著作權專區」。
若尚有任何專利申請與著作權等相關問題，歡迎洽詢本校全球產學營運及推廣處技術移轉中心，分機2651。
3. 授權書一式兩份，將簽署後論文公開授權書正本裝訂於審定書之後，辦理畢業离校時，除繳交一本論文至圖書館資訊處外，另一本繳交至教務處註冊課務組。

• **電子論文** 此項授權同意以非專屬、無償方式授權予本校圖書館，不限地域、時間與次數，以微縮、光碟或數位化方式將論文全文(含摘要)進行重製、及公開傳輸，亦提供讀者非營利使用線上檢索、閱讀、下載或列印。

立即公開 傳輸數位檔案

因特殊原因，校內 **讀於3年後** 公開，校外(含國家圖書館) **讀於3年後** 公開或上載網路公開閱覽。

※ 電子論文延後公開原因：涉及機密。

延後公開理由：涉及機密。

※ 電子論文公開日期：校內 民國114年12月26日，校外(含國家圖書館) 民國114年12月26日

• **紙本論文** 此項授權同意以非專屬、無償方式授權予本校圖書館，不限地域、時間與次數，以紙本方式將論文全文(含摘要)進行收錄、重製與利用；於著作權法合理使用範圍內，讀者得進行閱覽或列印。

同意 **立即公開**

因特殊原因，**讀於3年後** 公開陳覽

※ 紙本論文延後公開原因：涉及機密。

延後公開理由：涉及機密。

※ 紙本論文公開日期：民國114年12月26日

學 號：D062620002
授 權 人：洪揚豪 (簽章)
洪揚豪

指導教授：張鼎張 (簽章)
張鼎張

中華民國 111 年 12 月 29 日

※ 此授權書嚴禁塗改

- 若欲修改權限，請登入系統修改後重新列印此授權書。
- 若論文定稿核准後欲異動授權書，請洽詢etd@mail.nsysu.edu.tw或校內分機2452。
- 授權書需自行列印兩份，請於圖書館和教務處辦理離校手續時，裝訂於繳交的紙本論文內。

致謝

時光匆匆、歲月如梭，六年多的博士生涯即將邁入尾聲，這一路走來雖有難過失意，卻也充實豐沛，感謝讓我成長且蛻變的一切點滴。萬分感謝我的指導教授張鼎張老師，提供了完善且優良的實驗室環境與專業知識的傳授，讓我在就讀博士班期間能無後顧之憂的研究，在老師的身上除了學習到半導體領域的專業知識外，也學到邏輯思考方式與尋找正確且關鍵資料的能力，老師對於研究的熱忱與待人處事態度使我在博士班期間深受影響並收穫良多，是老師使我人生翻轉，完成博士學位並進入一流公司就業。此外感謝口試委員台積電莫亦先經理、奈盾科技陳紀文董事長、海軍官校陳柏勳教授、中山大學蔡宗鳴教授撥冗審查，並在口試時給予指證與教導，使我的博士論文更加完善，特此感謝。

在博士班期間，感謝廖柏詠學長與蘇婉菁學姊給予我的指導，不論是初入實驗室時的基本知識的建立，或是後期的機制釐清與討論，甚至是對於待人處事的提醒與啟發。此外感謝實驗室的學長姐們日謙、建佑、柏瑋、崇巽、孝承、冠甫、擋車、錫紋、俞慶、懿霆、志陽、馨平、智程、皓軒在實驗上的指導與關心，也很感謝陳宏誌學長在博士班後期，協助並教導我計畫上的執行，學長做事的態度使我十分欽佩，是我的榜樣；也格外感謝陳柏勳學長，在論文撰寫和回稿的過程中給了我許多建議，在博班求學階段也多次與我促膝長談，為我指引正確的求學態度與方向；也感謝與我同時期的同學們福源、宇哲、豐閔、妤珊、穩仲、俊曲一起做報告與執行計畫，準備口試的這一年感謝有你們的協助與陪伴，希望你們未來的工作也都一帆風順。還要感謝實驗室的學弟妹們的實驗協助與討論，玉發、建傑、茂洲、笠荃、冠儒、奕年、娟瑋、育霖、昱安、佳撰、家宏、沛君，謝謝你們在 TFT 計畫上的協助執行與討論，有你們計畫才得以順利完成，期許你們未來一切順利，保持初心並持續向前邁進。

最後我要感謝我的家人，我的父親洪彰先生以及在天上的母親陳寶珍女士，有您們的養育與支持，我才能順利完成博士學位，也感謝我的哥哥洪揚智給予我生活上的支持與幫助，也特別感謝國防工業發展基金會提供獎學金，使我在博班生涯能無經濟後顧之憂的致力於研究。

謝謝各位出現在我的生命中，一路走來受到太多貴人的幫助無法一一細數，謹以此論文獻給所以幫助過、關心過我的人們，我會在人生的下個階段繼續發光發熱，謝謝你們。

洪揚豪 謹識

2023.01 中山大學

摘要

近年來，隨著科技不斷的演進，臺灣光電顯示與半導體科技產業的蓬勃發展，各類平面顯示器相關應用已成為生活周遭不可或缺的消費性電子產品，像是穿戴式電子顯示器(apple watch)與需高電流驅動的有機發光二極體(OLED)顯示器。而顯示器中又以薄膜電晶體(TFT)最為重要，其扮演著畫素開關以及調變亮度的角色；顯示器的好壞可由薄膜電晶體作為判斷的依據。

薄膜電晶體中又以銦鎵鋅氧(InGaZnO)作為主動層材料最受關注，與傳統的非晶矽(a-Si)與低溫多晶矽(LTPS)相比，非晶態下依然能維持良好的電子雲重疊，使其同時兼具良好的均勻性(uniformity)與載子遷移率(mobility)，且較寬的電子能隙(3.2eV)使其具有低漏電特性，有助於降低功率的損耗，然而其對於光與電場的可靠度問題仍待研究與解決。

本論文首先研究不對稱結構 InGaZnO 薄膜電晶體於照光與熱載子效應(Hot-carriers effect)下的劣化機制與物理模型。電流—電壓(I_D-V_G)與電容—電壓(C-V)曲線在照光與電應力的操作後均觀察到駝峰效應(hump effect)，這是由於不對稱電極導致電場分佈不均而後造成的載子注入行為，最後透過模擬與交流操作(AC operation)做進一步的物理模型驗證。

此外，為了應對可撓與可摺疊柔性顯示器面板的需求，與 InGaZnO 相比，以有機材料製作而成有機薄膜電晶體則更有潛力與發展性。有機材料是由溶液製程所製備，可使用旋轉塗佈(Spin coating)、噴墨(Ink-jet Printing)等製程製作；具有成

本低廉、易於大面積成膜、耐衝擊、製程溫度低可製作於可撓性基板上等特性。使得有機半導體在科技與產業發展上具有極大的吸引力。然而，有機材料易受環境氣氛以及照光的影響，可靠度與性能仍有待改善。

第二部分探討前瞻有機薄膜電晶體的傳輸模型，元件半導體層是使用新穎垂直相態分層所製備而成，與傳統有機薄膜電晶體相比，具有較高的均勻性與載子遷移率。透過量測不同溫度下的 I_D - V_G 曲線萃取活化能，並歸類其物理機制。最後得出結論為活化能的變化率是由於小分子官能基團不穩定的 π - π 鍵結所導致。

此外，有機半導體容易受到照光以及氣氛的影響。第三部分為探討垂直分層有機薄膜電晶體於可見光後造成的劣化行為，透過雙閘極結構的元件量測，推論出照光所造成的電子注入於高分子層，導致元件閥值電壓右飄，最後透過無高分子層元件驗證其物理機制。

第四部份研究有機薄膜電晶體於熱載子效應下的劣化機制，元件受大汲極電壓影響，有 DIBL 效應的發生；且在熱載子電應力下觀察到開態電流下降及閥值電壓左飄的劣化。最後透過使用終端孔洞結構能有效抑制其劣化，改善元件熱載子以及 DIBL 可靠度。

關鍵字: 薄膜電晶體、有機薄膜電晶體、銻鎳鋅氧、熱載子效應、傳輸機制、終端結構

Abstract

Recently, with the continuous evolution of technology and the vigorous development of optoelectronic display and semiconductor technology industries in Taiwan, applications of various flat-panel displays have become indispensable consumer electronic products in our daily life, such as wearable electronic displays (apple watch) and high current-drive organic light-emitting diode (OLED) displays. Thin-film transistors (TFTs), which plays the role in switching pixel and brightness modulation, are the most important in the development of displays. In addition, the quality of displays can be determined by the TFTs.

Indium allium zinc oxide (InGaZnO) has attracted the most attention as the active layer material among TFTs. In contrast to traditional amorphous silicon (a-Si) and low temperature polysilicon (LTPS), a-InGaZnO contains the advantages of both. The overlap of effective electronic cloud in the amorphous state enables InGaZnO to remain better mobility than a-Si, and better uniformity than LTSP due to the grain boundary free. The wider energy gap of InGaZnO (3.2eV) suppresses the Band-to-Band-Tunneling (BTBT), which presents the low leakage current. However, the reliability of illumination and that of the electric field on InGaZnO still need to be investigated and solved.

In this dissertation, the degradation mechanism and the physical model of asymmetric InGaZnO thin film transistors under illumination and the hot-carriers effect

are investigated first. The current-voltage (I_D - V_G) and capacitance-voltage (C-V) curves both show a hump effect after the operation of illumination and electrical stress. The hump effect is caused by the uneven distribution of the electric field induced by the asymmetric electrode. Finally, the simulation and the pulse modulation are implemented for the further physical model verification.

In addition, in order to meet the needs of flexible and foldable display panels, organic TFTs have more potential and development than InGaZnO. Organic materials are fabricated by the solution process produced by spin coating and ink-jet printing, which contains the advantages of low cost, large-area film formation and impact resistance. The low process temperature enables organic materials to be fabricated on flexible substrates. These advantages of organic semiconductors make it extremely attractive in technology and industrial development. However, organic materials are easily affected by ambient atmosphere and illumination, and its reliability and performance still need to be improved.

The second part of this dissertation discusses the transport model of the novel organic thin film transistor. The semiconductor layer of the device is prepared by using a novel vertical phase-separated method, which helps perform higher uniformity and carrier mobility that traditional organic thin film transistors are not capable of achieving. The activation energy is extracted by measuring the I_D - V_G curves at different temperatures, and the physical mechanism is classified. The variation of activation energy is attributed

to the unstable π - π bond of small molecular functional groups.

In addition, organic semiconductors are easily affected by light and atmosphere. The third part of this dissertation investigates the degradation behavior of the vertical phase separated organic TFT after light illumination. It is inferred regarding the degradation that the electrons generated by illumination are injected into the polymer layer, which cause the device threshold voltage to shift positively. The physical mechanism is verified by the device without the polymer layer.

The fourth part studies the degradation mechanism of organic TFTs under the hot carrier effect. There is a DIBL effect occurring in the device at the high drain voltage. In addition, an on-state current drop and a negative threshold voltage shift are observed under the hot-carrier stress (HCS). The deterioration can be effectively suppressed by using the terminal-via structure, and the device hot carrier and DIBL reliability can be improved.

Keywords: Thin-film Transistors (TFTs), Organic Thin-film Transistors (OTFTs), Indium gallium zinc oxide (InGaZnO), Hot carrier effect, Transport mechanism, terminal structure

Contents

論文審定書	i
摘要	iii
Abstract	vii
Contents	x
Figure Captions.....	xiii
Table Captions	xix
Chapter 1 Introduction	1
1.1 Overview of Active-Matrix Flat Panel Displays	1
1.2 Overview of Metal Oxide Semiconductors	3
Reference	6
Chapter 2 Literature Review of Organic Thin-film Transistors	13
2.1 Introduction of Organic Thin-film Transistors	13
2.2 Types of Organic Materials	13
2.3 Self-assembled monolayer (SAM)	14
2.4 Transport Mechanism of Organic Materials.....	14
2.4.1 Band Transport	16
In the band transport model, the carrier mobility exponentially declines with the temperature increasing due to the phonon scattering. Normally, the band transport is found in inorganic semiconductors. According to J.H. Schön et al [2.13], the mobility decreases with temperature increasing in single crystal pentacene, as shown in Figure 2-6. However, the fabrication of crystalline state organic semiconductors has no advantages in terms of panel market applications, which is rarely discussed.	
.....	16

2.4.2 Hopping Transport.....	16
2.4.3 Multiple Trapping and Release (MTR) Model.....	16
Reference	18
Chapter 3 Parameter Extraction	25
3.1 The V_T extraction method.....	25
3.2 The subthreshold swing extraction method	27
3.3 The carrier mobility method.....	27
Chapter 4 Hump Effect on Asymmetric InGaZnO TFT	30
4.1 Introduction	30
4.2 Experimental architecture.....	31
4.3 Result and Discussion.....	32
4.4 Summary.....	38
Reference	39
Chapter 5 Transfer Mechanism on Phase Separated OTFT	48
5.1 Introduction	48
5.2 Experimental architecture.....	49
5.3 Result and Discussion.....	52
5.4 Summary.....	56
Reference	58
Chapter 6 Light induce V_T degradation on phase separated OTFT.....	68
6.1 Introduction	68
6.2 Experimental architecture.....	69
6.3 Result and Discussion.....	69
6.4 Summary.....	73
Reference	74

Chapter 7 Improving DIBL and HCS Reliability on OTFT by Terminal-via.....	84
7.1 Introduction	84
7.2 Experimental architecture.....	85
7.3 Result and Discussion.....	87
7.4 Summary.....	91
Reference	93
Chapter 8 Conclusion	104

Figure Captions

Chapter 1

Figure 1-1. The evolution and application of displays	8
Figure 1-2. The required mobility and number of pixels for high resolution displays.	8
Figure 1-3. The schematic and simplified circuit diagram of AM-LCD.	9
Figure 1-4. The schematic and simplified circuit diagram of AM-OLED.	9
Figure 1-5. The crystalline state of ZnO, Ga ₂ O ₃ , and In ₂ O ₃	10
Figure 1-6. The Hall mobility of ZnO, Ga ₂ O ₃ , and In ₂ O ₃	10
Figure 1- 7. Comparison of various TFT active materials.....	11
Figure 1- 8. Comparison of the crystalline and amorphous state between Si and InGaZnO.....	11

Chapter 2

Figure 2-1. The application of organic TFTs.	20
Figure 2-2. two types of organic semiconductor materials, organic polymers and small molecules.	20
Figure 2-3. The comparison between Pentacene and TMTES-Pentacene.....	21
Figure 2-4. The schematic diagram of the work function modulation with and without SAM layer.	21
Figure 2-5. The formation of conducting properties of organic material, and the σ bond and π bond.	22
Figure 2-6. The relationship between temperature and mobility in single crystal Pentacene.	22

Figure 2-7. The formula and schematic diagram of hopping model in small molecule organic material. 23

Figure 2-8. Different defect state induced the variation of mobility, include band transport and multiple trapping and release model..... 24

Chapter 3

Figure 3-1. Multifunctional semiconductor measurement and analysis system 29

Figure 3-2. Low-temperature semiconductor measurement platform. 29

Chapter 4

Figure 4-1. Three-dimensional diagram of IGZO device with cross-section view, top view, and optical microscope image..... 42

Figure 4-2. Electron trapping behavior in via-type InGaZnO under HCS. 42

Figure 4-3. Cross-sectional schematic diagram of the a-InGaZnO TFT used in this study under hot carrier ($V_G= 15$ V, $V_D= 30$ V) and blue light illumination..... 43

Figure 4-4. I_D - V_G characteristics of the U-shaped drain under hot carrier stress and blue light illumination with various stress times from 0 to 1000s in (a) forward and (b) reversed operation. 43

Figure 4-5. Vertical energy band diagram near drain electrode with InGaZnO device under HCS and blue light illumination. 44

Figure 4-6. Silvaco TCAD electric field simulation of device used in this dissertation under hot carrier stress. 44

Figure 4-7. COMSOL simulated electric field distribution with three-

dimensional geometry under HCS, and corresponding trapping behavior schematic diagram	45
Figure 4-8. Comparison of I_D - V_G curves between (a) simulated and (b) experimental data.....	45
Figure 4-9. (a) Drain capacitance-gate voltage (C_{GD}) and (b) source capacitance-gate voltage (C_{GS}) curves after hot carrier and illuminated stress.	46
Figure 4-10. Degradation behavior on (a) C_{GD} curve with corresponding trapping location at (b) corner and (c) flat.....	46
Figure 4-11. C-V curves of InGaZnO device after HCS (a) without and (b) with blue light.....	47
Figure 4-12. C-V curves of InGaZnO device under drain (a) DC and (b) AC pulse HCS. (c) Corresponding AC pulse operated duration. (d)	47

Chapter 5

Figure 5-1 The cross-section view of the schematic diagram and SEM image of the OTFT device.....	62
Figure 5-2. The phase separation of the organic semiconductor used in this dissertation.....	62
Figure 5-3. The I_D - V_G curves of OTFT device on (a) linear and (b) saturated operation with corresponding carrier mobility ($1.61 \text{ cm}^2/(\text{V}\cdot\text{s})$) and on/off current ratio (10^9), respectively.....	63
Figure 5-4. The I_d - V_d curves of OTFT device with V_g value from 5, 0, -5, -10, -15, and -20 V.	63
Figure 5-5. The (a) log and (b) linear I_D - V_G curves of OTFT device under various temperature region (108 ~ 348 K).	64

Figure 5-6. The (a) $\log(I_D)$ - V_G and (b) I_D - V_G curves measured under temperature region from 108K to 348K with fixed V_T	64
Figure 5-7. The $\ln(\mu) - 1/kT$ curves used to extract the activation energy of semiconductor material.....	65
Figure 5-8. (a) $\ln(\mu) - 1/kT$ curve separates into three regions. (b) region I, (c) region II, and (d) region III with E_a of 45, 21.5, and 3 meV, respectively, for corresponding temperature intervals.....	65
Figure 5-9. The curves of activation energy with different temperature interval.	66
Figure 5-10. Three dimensional bonding schematic diagram of TMTES-Pentacene and different bonding type of functionalized groups which present different defect states.	67

Chapter 6

Figure 6-1. Schematic diagram of device structure with top gate (TG) and dual gate (DG) device.....	76
Figure 6-2. The stress condition of the device under blue light illumination. 76	
Figure 6-3. The I_D - V_G curves of the TG device after illumination stress for 500 seconds.	77
Figure 6-4. The operated measurement of the DG device.....	77
Figure 6-5. The I_D - V_G curves of the DG device under various bottom gate voltages.....	78
Figure 6-6. The schematic diagram of the formation of bulk leakage under different bottom gate voltage with DG device.	78
Figure 6-7. The I_D - V_G curves of the DG device after blue light illumination with	

bottom gate voltage (a) 8, (b) 4, (c) 0, (d) -4, and (e) -8 V.....	79
Figure 6-8. Curve of $\Delta V_{th} - V_{BG}$	80
Figure 6-9. The vertical energy band diagram of the DG device during the device under situation of (a) blue light induced electron hole pairs, (b) electron trapping to polymer and base layer, and (c) bottom gate bias detrapping electrons.....	81
Figure 6-10. The influence of electron trapping location on the DG device..	82
Figure 6-11. The V_T fixed I_D - V_D curves of the DG device after blue light illumination with bottom gate voltage reaches at (a) 8 and (b) -8 V.	82
Figure 6-12. I_D - V_G curves of the polymer free device after blue light illumination for 500 seconds.	83
Figure 6-13. Annealing induce electron detrapping and corresponding I_D - V_G curves of 5 devices after annealing.....	83

Chapter 7

Figure 7-1. OTFT device with cross-section view schematic diagram and the image under optical microscope.....	96
Figure 7-2. Three-dimensional structure diagram and OM image of the terminal via device used in this dissertation.	96
Figure 7-3. I_D - V_G curves of standard device with channel length 15 μm under various drain voltage.	97
Figure 7-4. I_D - V_G curves of standard device with channel length 5 μm under various drain voltage.	97
Figure 7-5. I_D - V_G curves of terminal via device with channel length (a) 15 and (b) 5 μm under various drain voltage.	98

Figure 7-6. (a) The breakdown optical image of standard device under -40 V drain bias and (b) corresponding degradation behavior of hump effect. 98

Figure 7-7. Drain induced source barrier lowering (DIBL) effect. 99

Figure 7-8. The COMSOL electric field simulation under a -40 V drain bias of (a) standard and (b) terminal via structures. 99

Figure 7-9. COMSOL horizontal electric field simulation of the device with (a) standard and (b) terminal via structure. 100

Figure 7-10. I_D - V_G curves of (a) standard and (b) terminal via device after HCS for 1000 seconds. 100

Figure 7-11. The hole trapping behavior on the device schematic and vertical energy band diagrams after HCS. 101

Figure 7-12. I_D - V_G curves of (a) standard and (b) terminal via device with threshold voltage correlation after HCS for 1000 seconds. 101

Figure 7-13. The thermal conductivity of standard (organic material) and terminal via (SiO_2) device. 102

Figure 7-14. The model of I_{ON} degradation after HCS. 102

Figure 7-15. COMSOL 3-dimensional thermal field distribution of (a) standard and (b) terminal via device. 103

Table Captions

Table 3-1 Parameter definition and unit table	26
---	----