



國立中山大學材料與光電科學學系

博士論文

Department of Materials and Optoelectronic Science

National Sun Yat-sen University

Doctoral Dissertation

可撓式低溫多晶矽及有機薄膜電晶體開發

與物理機制探討

Development and Physical Mechanisms Establishment of

Flexible Low Temperature Polycrystalline Silicon and

Organic Thin Film Transistors

研究生：鄭宇哲

Yu-Zhe Zheng

指導教授：蔡宗鳴 博士

Dr. Tsung-Ming Tsai

中華民國112年1月

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Development and Physical Mechanisms Establishment of Flexible Low
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於中華民國 112 年 1 月 7 日經本委員會審查並舉行口試，符合博士學位論文標準。

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指導教授(蔡宗鳴) 蔡宗鳴 (簽名)

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鄭宇哲 謹識

摘要

近年來，柔性薄膜電晶體的應用範圍越來越廣泛，導致許多研發及研究人員投入更多的心力去開發相關的顯示器產品。隨著市場上的需求，智慧型手機從原本的平面式顯示、固定曲率邊界到可以重複彎曲摺疊的智慧型手機，柔性螢幕成為了現在及未來的顯示器趨勢。然而從柔性薄膜電晶體的彎曲的耐久性與電性的可靠度仍然有進步的空間。本論文探討可撓式低溫多晶矽薄膜電晶體在機械應力彎曲後彎曲耐久性的議題以及討論可撓式有機薄膜電晶體在彎曲後的物理機制並研究在指叉結構下的有機薄膜電晶體的電性劣化。

本博士論文第一章節主要討論低溫多晶矽薄膜電晶體在可撓式聚醯亞胺(Polyimide)基板上藉由改善氧化層品質來提升機械應力的耐久性。現階段的可撓式薄膜電晶體多半使用聚醯亞胺作為基板，除了具有可彎曲性外，且具有良好的熱穩定性以及化學特性。此外聚醯亞胺這材料可以整合於溶液製程，因此可以用旋轉塗布的方式進行沉積。沉積在可撓式基板的低溫多晶矽薄膜電晶體在經過多次彎曲過後，會因為材料之間楊氏係數的差異，使應力集中在氧化層的邊界處，造成 LTPS TFTs 的電性劣化，因此氧化層的品質對於 LTPS TFTs 來說是一個重要的關鍵。本章節中，研究使用化學氣象沉積(PECVD)沉積閘極氧化層時，通入不同氮濃度來提升二氧化矽的品質，以此增強低溫多晶矽薄膜電晶體的電性可靠度和機械應力的耐久性。藉由參數的萃取，如次臨界擺幅、載子遷移率、電性量測及機械應力測試等來調查氮氣對於 PECVD 在沉積氧化成時的影響，最後也利用電子束顯微鏡中的 X 射線能量散布分析儀來分析元素之間的差異。

本論文第二章節中，研究了在一系列不同氣氛中進行機械彎曲應力後有機薄膜電晶體的電性變化。在大氣環境中，經過 100,000 次通道寬度軸壓應力彎曲和彎曲半徑為 $R=5\text{ mm}$ 的長時間固定曲率後，可以看到臨界電壓 (V_T) 有嚴重的劣化行為。在彎曲後退火之後，對於動態和靜態彎曲測試， V_T 恢復到比初始 V_T

更負的值。在真空中進行彎曲測試，消除了環境大氣對 OTFT 的影響，表明器件在大氣彎曲下的退化主要由兩種機制決定。依次引入不同的氣體（濕度、氮氣、二氧化碳、氧氣）以了解它們對 OTFT 的影響。通過研究真空和大氣中的影響，我們能夠將機械彎曲對 OTFT 的物理影響與暴露於環境因素引起的變化區分開來。由於不同的機械彎曲條件，主動層表面的小分子會引起不同的電性特性。最後，提出了有機 TFT 的機械彎曲和大氣因素的物理模型。

第三部分調查了高電流操作下焦耳加熱對指插結構的有機薄膜電晶體 (OTFTs) 的影響。討論了具有不同指叉數量 ($N=4$ 、 6 和 10) 的元件之電性劣化。由於運行期間的熱量積累，在大電流下觀察到顯著的電性退化。在高電流及電壓的操作下，隨著指叉數量的增加，電性呈現不同程度的劣化，在 10 個指叉數量的元件中觀察到異常駝峰。為了驗證自熱效應對於 OTFTs 的影響，將 OTFTs 製備在不同導熱係數的材料上，聚酰亞胺和玻璃兩種基板，並在室溫和低溫下研究了電性劣化的原因。此外，還提出了交流操作來改變元件的加熱與散熱時間進而得到驗證。最後，透過了 Silvaco-TCAD 仿真來模擬元件的自熱效應，並說明手指數量對偏置應力的影響，並且提出劣化的原因。

關鍵字：可撓式電子元件、低溫多晶矽薄膜電晶體、有機薄膜電晶體、真空彎曲量測、氣氛量測、跳躍距離、指叉結構、自熱效應

Abstract

Recently, the application of flexible thin-film transistors has become more and more striking, which leading many R&D and researchers to put more effort into developing related display products. With the demand in the market of display, smart phones have evolved from the traditional flat display to the fixed curvature boundary and the smart phone that can be repeatedly bent and folded. However, there is still room for improvement in the bending endurance and electrical reliability of the flexible thin film transistor. The topics discussed in this dissertation include the bending endurance of flexible low temperature polysilicon thin film transistors under mechanical bending stress, the physical mechanism of flexible organic thin film transistors after bending, and studies the electrical deterioration of organic thin film transistors with multi-Finger structure.

The first part of this dissertation discusses low-temperature polysilicon thin film transistors on flexible polyimide (Polyimide) substrates to improve the endurance of mechanical stress by improving the quality of the oxide layer. Most of the current flexible thin film transistors use polyimide as the substrate, which not only has flexibility but also has good thermal stability and chemical properties. In addition, the polyimide can be integrated with the solution process, so it can be deposited by spin coating. However, due to the difference Young's moduli between deposited layers, the result of the low-temperature polysilicon thin film transistor with the flexible substrate shows the strain will be concentrated at the edge of the gate insulator under the mechanical bending stress, that cause the electrical deterioration of the LTPS TFTs. Therefore, the quality of the oxide layer is an important key for LTPS TFTs.

In this chapter, the use of chemical vapor deposition (PECVD) to deposit the gate oxide layer is studied to improve the quality of silicon dioxide by introducing different

helium concentrations, thereby enhancing the electrical reliability and mechanical stress endurance of low-temperature polysilicon thin film transistors. Through the extraction of parameters to investigate the effect of different helium concentrations on PECVD deposition the gate insulator, such as subthreshold swing, carrier mobility, electrical measurement, and mechanical stress test. Finally, transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDS) to analyze element changes between materials

In the second part, the electrical characteristics of organic thin film transistors after mechanical bending stress in a series of different atmospheres are investigated. In atmospheric environments, severe degradation behavior of the threshold voltage (V_T) can be observed after 100,000 channel width axial compressive stress bends and a long-term fixed curved with a bending radius of $R=5$ mm. Following annealing after bending, V_T recovered to a more negative value than the initial V_T for both dynamic and fixed curved bending. Performing bending tests in a vacuum, which eliminates the influence of the ambient atmosphere on the OTFTs, shows that the degradation of the device under atmospheric bending is dominated by two mechanisms. Sequentially introduce different gases (humidity, nitrogen, carbon dioxide, oxygen) to clarify their effect on OTFTs. According to the experiments described above, the effects in the vacuum and atmosphere, we are able to separate the physical effects of mechanical bending on the OTFTs from the changes induced by exposure to environmental factors. Due to different mechanical bending conditions, small molecules on the surface of the active layer can induce different electrical properties. Finally, physical models of mechanical bending and atmospheric factors of organic TFTs are proposed.

The third section investigates the effect of Joule heating on organic thin-film transistors (OTFTs) with multi-finger structured under high-current operation. The electrical degradation of OTFTs with different numbers of fingers ($N = 4, 6$ and 10) is

discussed. Significant electrical degradation was observed at high current due to heat accumulation during operation. Under the operation of high current and voltage, with the increase of the number of fingers, the electrical characteristics show different degrees of deterioration, and an abnormal hump is observed in the OTFTs with the $N=10$. In order to verify the influence of self-heating effect on OTFTs, OTFTs were fabricated on materials with different thermal conductivity, polyimide and glass substrates, and the causes of degradation were investigated at both room temperature and low temperatures. In addition, AC operation is proposed to change the heating and cooling time. Finally, Silvaco TCAD simulation is used to simulate the self-heating effect of the OTFTs devices, and the effect of the number of fingers on the bias stress is explained, and the reason for the deterioration is proposed.

Index Terms: Flexible electronic devices, Low temperature polycrystalline silicon TFT, Organic TFT, Vacuum bending measurement, Atmosphere measurement, Hopping distance, Multi-finger structure, Self-heating effect.

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Fig. 5-3 (a) The electrical transfer curves of $N=10$ OTFT fabricated on PI substrate during stress and (b) OTFT fabricated on glass substrate stress at 200 K. (c) Schematic diagram of OTFTs on glass substrate during AC stress condition (d) Waveforms used to operate the OTFTs (e) OTFT electrical transfer curves under bias stress of 40 μ s on state pulse and (f) under 10 μ s on state pulse. 82

Fig. 5-4 Silvaco TACD thermal simulation of bias stress and heat distribution for $N=4$, $N=6$ and $N=10$ devices. 83

Fig. 5-5 (a) Top view of multi-finger structure OTFT and electrical degradation was divided into central and side region. (b) the abnormal hump of I_D - V_G corresponded to central and side region. (c) and (d) shows the different amount of energy band lift, and the schematic diagram of different electron injection in base layer, respectively. 83

Table Caption

Table 4-1. Young's modulus of OTFT each material.68