1
Introduction

Organic semiconductor technology has attracted considerable research interest in view of its great promise for large area, low-end, lightweight, and flexible electronics applications [1]. Owing to their processability advantages and unique physical (i.e., electrical, optical, thermal, and magnetic) properties, organic semiconductors can bring exciting new opportunities for broad-impact applications requiring large-area coverage, mechanical flexibility, low-temperature processing, and low cost. Thus, organic semiconductors have appeal for a broad range of devices including transistors, diodes, sensors, solar cells, and light-emitting devices. Figure 1.1 depicts a number of application domains that can benefit from the versatility of organic electronics technology [2]. Since their proof of concept in the 1980s, the impressive development in organic semiconductor materials has led to performance properties that are competitive with amorphous silicon (a-Si), increasing their suitability for commercial applications [3].

The transistor is a fundamental building block for all modern electronics; transistors based on organic semiconductors as the active layer are referred to as organic thin film transistors (OTFTs). A number of commercial opportunities have been identified for OTFTs, including flat panel active-matrix liquid crystal displays (LCDs) or active matrix organic light-emitting diode displays (AMOLEDs), electronic paper (e-paper), low-end data storage such as smart cards, radio-frequency identification (RFID) and tracking devices, low-cost disposable electronic products, and sensor arrays; more applications continue to evolve as the technology matures [4]. Figure 1.2 illustrates a few commercial opportunities envisioned for OTFTs.

The unique features which give organic electronics a technological edge are simpler fabrication methods and the ability to mechanically flex. Fabrication of organic electronics can be done using relatively simple processes such as evaporation, spin-coating, and printing, which do not require high-end clean room laboratories. For example, solution-processable organic thin films can be deposited by spin coating, enabling fast and inexpensive coverage over large areas. Inkjet printing techniques can be used to deposit soluble organic inks. In addition, low-temperature processing and the mechanical flexibility of organic materials make them highly favorable for implementation on robust substrates

Organic Thin Film Transistor Integration: A Hybrid Approach, First Edition. Flora M. Li, Arokia Nathan, Yiliang Wu, and Beng S. Ong.

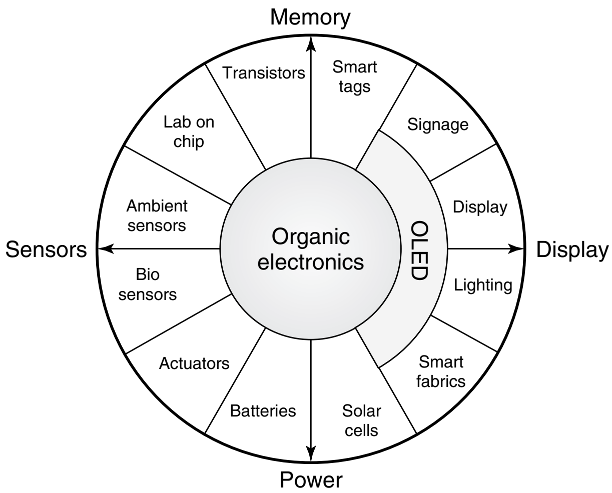

Figure 1.1 A broad range of products and technologies inspired by organic electronics [2].

Figure 1.2 Examples of commercial opportunities for OTFTs.

with non-conventional form factors. In general, organic electronic devices are not expected to compete with silicon devices in high-end products, because of their lower speed as compared to silicon. Thus organic electronics is intended to complement conventional silicon technology. It is expected to thrive in a different market domain targeting lower resolution, cost-effective mass production items such as identification tags, smart cards, and pixel drivers for display and sensor technology.

1.1
Organic Electronics: History and Market Opportunities

Historically, organic materials (or plastics) were viewed as insulators, with applications commonly seen in inactive packaging, coating, containers, moldings, and so on. Research on the electrical behavior of organic materials commenced in the 1960s [5]. Photoconductive organic materials were discovered in the 1970s and were used in xerographic sensors. The announcement of conductive polymers in the late 1970s [6], and of conjugated semiconductors and photoemission polymers in the 1980s [7], gave new impulse to the activity in the field of organic electronics. Polyacetylene was one of the first polymers reported to be capable of conducting electricity [8], and it was discovered that oxidative doping with iodine causes the conductivity to increase by 12 orders of magnitude [9]. This discovery and the development of highly-conductive organic polymers was credited to Alan J. Heeger, Alan G. MacDiarmid, and Hideki Shirakawa, who were jointly awarded the Nobel Prize in Chemistry in 2000 for their 1977 discovery and development of oxidized, iodine-doped polyacetylene.

The continued evolution of organic semiconductor materials from the standpoint of electrical stability, processability, functionality, and performance is enabling realization of high-performance devices in laboratory environments [10–14]. The advancement in organic semiconductor materials is starting to prompt the transition of the technology from an academic research environment to industrial research and development (R&D). The shift toward industrial R&D is aided by the establishment of several government-sponsored research initiatives [15, 16], the founding of various organic electronics driven associations and companies [17–19], and the development of IEEE standards for the testing of organic electronics devices [20]. The increased cooperative efforts between academia, industry, and government are vital to the development of a strong materials and manufacturing infrastructure [21–26].

The outlook for low-cost production of organic electronics is a key driver for market opportunities in this area. To achieve these cost targets, low-cost materials, cost-effective processes, and high-volume manufacturing infrastructure are required. The development of high-volume roll-to-roll manufacturing platforms for fabrication of organic circuits on continuous, flexible, low-cost substrates, has been reported. These platforms are based on the integration of lithography, vacuum deposition, and printing technologies. It has been forecast that an organic semiconductor fabrication facility can be built for far less than the cost of a silicon semiconductor fabrication facility [3]. The high cost of silicon-based foundries can be attributed to the sophisticated wafer processing and handling equipment, high-resolution lithography tools, wafer testing equipment, clean-room environment, and costly chemical distribution and disposal facilities. In contrast, the cost reduction forecast for an organic electronic manufacturing facility is expected to be derived from lower materials cost, less sophisticated equipment, simpler manufacturing technologies, less stringent demands on clean-room settings, and reduced

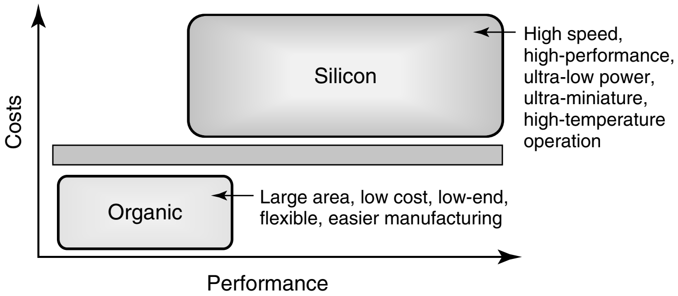

Figure 1.3 Illustration of cost versus performance comparison of silicon technology and organic semiconductor technology.

waste output. However, the potential savings in the manufacturing cost of organic electronics come with the trade-off of lower performance.

Figure 1.3 provides a conceptual view of the cost-and-performance sectors served by silicon technology and organic semiconductor technology. It must be noted that organic semiconductor devices do not offer the same electrical performance as silicon devices. While silicon technology is aimed for high-end, high performance, and high processing power electronic products, organic semiconductor technology appeals for lower-end, cost-effective disposable electronics products.

One of the most frequently discussed opportunities for organic electronics is their integration as the driver backplane of flexible displays. Specifically, printed organic semiconductor materials are strong candidates for novel electrically active display media. The same applies to radio frequency interrogation devices. An overview of these OTFT-based applications and their current market status is presented next. Note that, at present, a-Si thin film transistors (TFTs) and polycrystalline silicon (poly-Si) TFTs are the key backplane technologies used in flat panel display products. Therefore, OTFTs are not intended to displace a-Si TFTs in large-area high-resolution flat panel displays. Instead, they will have a bigger impact on lower-cost flexible displays and e-paper applications. The key features of OTFT and a-Si TFT are compared in Table 1.1.

1.1.1
Large-Area Displays

The application of OTFTs for large area displays has been demonstrated by a number of companies and research institutions. For example, Plastic Logic Ltd. demonstrated the integration of an OTFT-driven backplane to a Gyricon display in 2003 [27]. The active-matrix display backplane was inkjet printed and drove a 3000-pixel display that was fabricated on glass. In early 2007, the world's first factory was built to produce plastic electronic devices [18].

A number of corporations have also invested in R&D for OTFT-driven large area displays. Examples include Sony, Samsung, Kodak, LG Philips, Motorola, 3M, and

Table 1.1 Comparison of OTFTs and amorphous silicon (a-Si) TFTs.

	OTFT	**a-Si TFT**
Material	Organic semiconductor as active layer; p-type, n-type, ambipolar	a-Si as active layer n-type
Processing	Spin-coat, print, evaporation. Low temperature (e.g., room temperature)	Plasma enhanced chemical vapor deposition (PECVD) $T < 350\,^{\circ}\mathrm{C}$
Mobility	Can be comparable to a-Si	$\sim 1\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$
Substrate and form factor	Variety of substrates and flexible form factor Mechanically flexible	Glass (most common), Plastic (in development)
Mechanical flexibility	Bendable	Fragile and brittle
Electrical stability	Rapid degradation, but degradation stabilizes (may be favorable for devices that turn on for a longer time)	Slower bias-induced degradation, but degradation does not stabilize
Pros	Potentially no clean-room, lower cost	More mature and stable
Cons	Process challenge; device performance, stability, and lifetime	Mechanical flexibility (stress) Higher processing temperature
Key applications	Numerous: displays, RFID tags, sensors, disposable electronics	Circuits for large-area displays and sensors array backplane
Outlook	New opportunities: smaller/flexible displays, disposable electronics, smart textiles	Continue to excel in AMLCD, AMOLED, active-matrix sensor technologies

Hewlett-Packard. In 2007, Sony demonstrated a 2.5 in. AMOLED display driven by OTFTs [28]; LG Philips' LCD Division presented a high resolution active-matrix liquid crystal display (AMLCD) with an OTFT-driven backplane fabricated using solution processing [29]; and Samsung Electronics reported an active-matrix display using printed OTFTs [30].

1.1.2 Rollable Displays

The mechanical flexibility of organic materials makes them particularly attractive for rollable or flexible displays. Polymer Vision, a spin off from Royal Philips Electronics, was a pioneer in demonstrating the capability of rollable displays, which were produced by combining ultrathin flexible OTFT-driven active-matrix backplane technology and flexible electronic ink (E-Ink) display technology. In January 2008, Polymer Vision introduced their first rollable display product, called *Readius*®, a pocket-sized device, combining a 5″ rollable display with high speed connectivity. The Readius® demonstrated a merger of the reading-friendly strengths of electronic-readers with the high mobility features of mobile phones, along with instant access to personalized news and information [31]. Demand for larger mobile displays is accelerating as telecom players push mobile content and mobile advertisements. The solution is to unroll the display when needed and simply store it away when not in use. Therefore, rollable display enabled devices are expected to be an emerging commodity for new generations of portable communication devices, thus presenting exciting commercial opportunities for OTFT-driven display backplane technology.

1.1.3
Radio Frequency Identification (RFID) Tag

One of the frequently promoted applications for organic electronics is the RFID tag. The RFID tag is a wireless form of automated identification technology that allows non-contact reading of data, making it effective for manufacturing, inventory, and transport environments where bar code labels are inadequate. Advantages of organic-based RFID tags over silicon-based tags include mechanical flexibility (e.g., bendable) and direct fabrication onto large area substrates using simple printable methods. The attractiveness of printed organic semiconductor materials and manufacturing platforms has drawn the involvement of several companies (e.g., 3M, Siemens), start-ups (e.g., OrganicID, ORFID Corp.), and research institutions to develop technology for organic-based RFID tags [25, 26]. For example, a 64-bit inductively-coupled passive RFID tag on a plastic substrate was demonstrated, operating at 13.56 MHz and with a read distance of over 10 cm. These specifications are approaching item-level tagging requirements, paving the way for low-cost high-volume production of RFID tags, with the potential to replace barcodes [19, 32, 33].

1.1.4
Technological Challenges

Organic electronics have reached early stages of commercial viability. Personal electronic devices incorporating small displays based on organic light-emitting diodes (OLEDs) are now available. However, many challenges still remain that are currently hindering the wide adoption of OTFTs in electronic devices. The shortcomings of OTFTs include limited charge carrier mobilities, high contact resistance, relatively higher operating voltages, device reliability issues (e.g., stability, shelf-life under operation), and limited availability of robust/mature patterning techniques and fabrication processes that are compatible with organic thin films. These technical challenges can be grouped into two categories: device performance and device manufacture.

1.1.4.1 Device Performance

One of the limitations of organic semiconductor materials, compared to silicon technology, is their intrinsically lower mobility. Most organic semiconductor thin films are composed of a mixture of polycrystalline and amorphous phases. The hopping process between molecules in the disordered regions often limits charge-carrier mobilities in organic semiconductor films [34]. To improve charge transport and device mobility, the disordered phase must be suppressed. The two most common approaches considered are:

- Tuning the molecular structure of the organic semiconductor during material preparation. Examples include modifying molecular parameters (e.g., regioregularity, molecular weight, side-chain length, doping level) during material synthesis, and altering processing conditions during material deposition (e.g., thermal annealing, solvent selection, film thickness, deposition methods, and parameters) [10–12].
- Exploiting interfacial phenomena to improve molecular ordering of the semiconductor layer during device processing. Examples of interfacial phenomena include semiconductor alignment using self-assembled monolayers (SAMs), surface-mediated molecular ordering, surface dipoles, physical alignment, and photoalignment [35]. Surface control using SAMs is a well-known technique for such interface modifications and can provide microscopically good interface regulations.

We will expand on the latter approach, where the interfaces are engineered to enhance device performance. An OTFT has two critical interfaces: the interface between the gate dielectric and the organic semiconductor, and the interface between the source/drain contacts and the organic semiconductor. The two interfaces dictate charge transport and charge injection in OTFTs, respectively, thus having an overriding influence on the device characteristics. We will investigate interface modification techniques for these two device interfaces, with an attempt to enhance device performance.

One of the areas of interest lies in the integration of a plasma-enhanced chemical vapor deposited (PECVD) gate dielectric with a solution-processable organic semiconductor for OTFT fabrication. Here, we study PECVD silicon nitride (SiN_x) as the gate dielectric. Past experiments have reported limited OTFT performance with a PECVD SiN_x gate dielectric, and have attributed this to surface roughness and unfriendly (or non-organic-friendly) interfaces of SiN_x. However, PECVD SiN_x as a gate dielectric has excellent dielectric properties. It is scalable to large areas and can be deposited at low temperatures, making it compatible with plastic substrates. We believe the critical factors to enable integration of SiN_x in organic electronics lie in identifying a suitable SiN_x composition and an agreeable interface modification process. Strategies to address these factors and to enable the use of SiN_x while delivering acceptable device performance remain the ultimate goal.

1.1.4.2 **Device Manufacture**

Since a major driving force behind OTFT technology is the manufacture of low-end, low cost, and disposable electronic devices, this demands a fabrication process that allows high volume production at low cost. Moreover, the process should be able to produce stand-alone devices, device arrays, and integrated circuits (ICs) of acceptable operating speed, functionality, reliability, and lifetime. However, an integration process that can meet the above requirements for production of high yield, stable OTFTs is, currently, non-existent.

Conventional photolithography processes for manufacturing silicon-based microelectronics are not completely amenable to organic electronics. Although photolithography has the advantage of producing high resolution, complex device structures with excellent precision, the process must be modified to ensure compatibility with organic materials. Advanced printing techniques (e.g., inkjet printing or nanoimprinting) that take advantage of the solution-processability of organic materials are favorable for achieving the goals of low-cost and high-volume production. Inkjet printing technology is particularly attractive because it offers the advantages of fast, direct imaging and single-step print processing, precise deposition of the organic ink only where it is needed (thus reducing waste), compatibility with flexible substrates, large area processing, and high material usage efficiency. However, because the requirements of printing electronic functions are very different from those of printing visual images, the adaptation of inkjet systems for processing organic electronic devices will require extensive optimization of printing parameters and processing conditions; in addition, technological concerns such as layer continuity and multilayer registration must be resolved. Inkjet printed organic devices with good performance have been demonstrated; however, low device yield is an issue. We will address the challenges in OTFT manufacture by exploring a hybrid manufacturing approach that combines a photolithography process with a novel inkjet printing technique. This delivers an integration strategy with workable manufacturing yields while lowering costs compared to conventional processes.

1.1.5
Scope and Organization

As the material properties and processing technology for organic electronics continue to advance and mature, the next phase of development is directed at integrating OTFTs into circuits and systems. It is not within the scope of this book to review organic semiconductor material development (which is the strength of chemical physicists), but rather to advance OTFT research from an engineering and integration perspective. By utilizing and assimilating existing materials, techniques and resources, we explore a number of approaches to deliver higher performance devices and demonstrate the feasibility of organic circuits for practical applications. The key focus areas include:

- Development of OTFT fabrication strategies to enable circuit integration (Chapter 3);

- Optimization of PECVD gate dielectric composition and structure to improve OTFT performance (Chapter 4);
- Investigation of interface engineering methodologies to enhance the dielectric/semiconductor interface and the contact/semiconductor interface (Chapters 5 and 6);
- Finally, the scientific and technical knowledge acquired from these investigations is applied to demonstrate the integration of OTFTs into functional circuits for active-matrix display and RFID applications (Chapter 7).

The results presented here stem primarily from research conducted at the Giga-to-Nano (G2N) Labs, University of Waterloo, in collaboration with the Xerox Research Centre of Canada (XRCC), which granted access to its high quality, high performance, stable organic semiconductor material. In particular, Xerox's solution-processable poly(3,3‴-dialkylquarterthiophene) (PQT-12) polymer semiconductor forms the basis for the majority of the OTFT experimental work [10] discussed in this book.

This book is divided into eight chapters. We begin with an introduction to organic electronics and market opportunities for OTFT technologies in Chapter 1. Chapter 2 examines the OTFT technology in greater depth, with a review of fundamental properties of organic semiconductors and a discussion of OTFT operation, device architectures, and material selection. Chapter 3 presents integration strategies to enable the fabrication of OTFT circuits.

With the aim of improving OTFT performance, optimization of PECVD gate dielectrics is explored in Chapter 4. Interface engineering strategies to improve charge transport by dielectric/semiconductor interface treatment methods and to enhance charge injection by contact/semiconductor interface modification techniques are given in Chapters 5 and 6, respectively. The objectives for these investigations are to enhance OTFT characteristics via functionalization of the gate dielectric material and the device interfaces, and to develop a better understanding of the materials and interfaces for OTFTs.

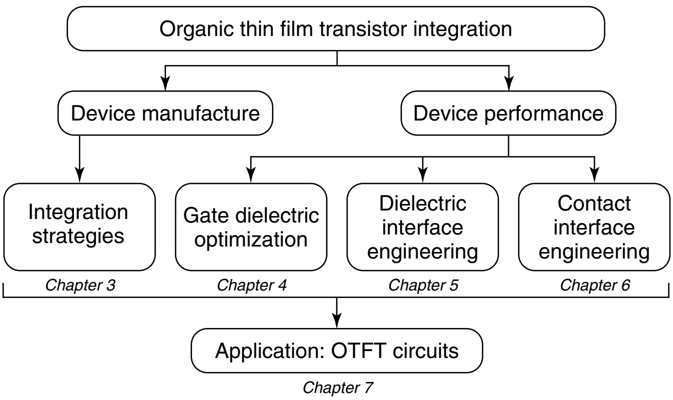

Figure 1.4 Illustration of the organizational structure of the book.

Chapter 7 demonstrates integration of OTFTs into functional circuits. Finally, Chapter 8 presents a summary of the outlook and future challenges related to OTFT integration. The structural design of the book is summarized in Figure 1.4, which illustrates the flow of the various topics related to advancing device manufacture, device performance, and OTFT circuit integration.

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