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# Solution-processable organic and hybrid gate dielectrics for printed electronics

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A B S T R A C T

Over the few past decades, printed electronics as an emerging technology have achieved tremendous progress in material design and device fabrication. OTFTs are the key building blocks of many electronic devices. Printable OTFTs have promising applications in flexible circuits, sensors and backplanes for active-matrix displays. Gate dielectrics play key roles in OTFTs to afford electrical insulating properties and interfaces for charge transport. In this paper, we review the recent progress of polymer and hybrid dielectrics for printable OTFTs. The requirement and mechanism of the gate dielectrics, different types of materials and remaining challenges for this field are presented. This review will provide comprehensive and timely guidelines for dielectric design and device engineering towards the fabrication of printed low-voltage and high-performance OTFTs.



**Contents**



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## 1. Introduction

Over the past two decades, printed electronics have been greatly progressed in materials and devices [1–4], which can be widely applied in many fields, such as flexible displays, wearable electronics, RFID tags, and disposable sensors, due to their low-temperature and solution processing, large-area fabrication, low cost, and compatibility with flexible substrates. Thin-film transistors (TFT) are the fundamental elements for printed electronics in constructing circuits and systems for signal processing. Organic TFTs (OTFTs), utilizing solution-processable organic semiconductor and dielectric materials, own several competitive advantages, including low-cost and high-throughput manufacturing and excellent intrinsic flexibility, and are compatible with arbitrary substrates, such as plastic films and papers, which enable easy hybrid integration of multifunctional materials [5–7]. Therefore, OTFTs have great promise for creating intelligent surfaces with flexible displays, sensors and other electronic functions for a wide range of ever-increasing electronics applications [8–10].

In recent years, significant efforts have been devoted to the research and development of high-performance polymer and small-molecule organic semiconductor (OSC) materials [11]. Extraordinarily high mobility values over 10 $cm^2 V^{-1} s^{-1}$ have been reported in the literatures for both p-type and n-type OSCs [12–17]. However, many of these high-mobility OTFT studies have used commercially available thermally oxidized $SiO_2$ as the gate dielectric with a treatment of self-assembled monolayer (SAM) to suppress the influence of hydroxyl groups on the OSC molecular packing and carrier transport. To achieve a competitive OTFT technology that can be incorporated into printing processes for circuit integration on a flexible substrate, solution-processable polymer and hybrid dielectrics are preferred because of their low-temperature processing and excellent mechanical flexibility. Thus, finding a suitable gate dielectric layer to match high-performance OSCs is vital to achieving printed OTFTs with ideal device performances, including high carrier mobility, large on/off ratio, low threshold voltage, steep subthreshold swing and good stability.

Previously, various solution-processable polymer dielectrics have been extensively studied for OTFTs [18–20]. However, most polymeric dielectrics have relatively low dielectric constants ($k$). As a result, the reported OTFTs often require operating voltages of a few tens of volts or even above 100 V since a thick dielectric layer is required to eliminate gate leakage and be compatible for large-area solution-based processing. Although high-$k$ dielectrics could help achieve a low operation voltage with a thicker layer, there are very few choices of suitable polymer dielectrics with sufficiently high $k$ values. Developing high-$k$ polymer dielectric compatible with OSCs has been one focus of OTFT studies but remains a key challenge [21]. Normally, when a high-$k$ gate dielectric layer is directly interfacing the OSC channel, the energetic disorder caused by the dipoles in the high-$k$ dielectric may tend to trap carriers from the gate-bias-induced conduction channel [22]. The resulting charge-carrier localization could cause not only mobility degradation but also increased hysteresis and device instability. Only few exceptional cases have been observed when a conductive channel is separated from the neighbouring high-$k$ gate dielectric by long enough side chains of polymers [21]. To address the mobility degradation and instability issues with OTFTs using high-$k$ polymer gate dielectric, a low-$k$/high-$k$ bilayer structure gate dielectric has been studied with a thin low-$k$ dielectric layer inserted between the high-$k$ dielectric and the OSC channel to suppress the dipole field effect [23,24]. In another case, hybrid dielectrics, including structures of inorganic/polymer composites and inorganic/polymer double layers, have been developed to obtain a large capacitance for a low operating voltage and good interface of semiconductor and dielectric for good carrier transport. A type of solid polymer electrolyte, so-called ion gels, has also been studied as a gate dielectric for low-voltage OTFTs. Ion gels are obtained by blending ionic liquids with a gelating triblock copolymer to form a physically crosslinked network that features a very large specific capacitance exceeding 1 $\mu F/cm^2$ with a thickness of approximately 1 $\mu m$, which can substantially reduce the operation voltage and simultaneously achieve very high driving currents [25]. Crosslinking of polymer dielectrics by photo or thermal reactions has been studied to enhance solvent resistance and improve the electrical robustness [19,20].

In this paper, we review the recent progress in polymer and hybrid dielectrics for high-performance OTFTs. This review presents the mechanisms and requirements of dielectrics for printable OTFTs, different types of printable gate dielectrics and the remaining challenges of dielectrics in printed OTFTs. This review provides important guidelines for materials design and device engineering towards fabricating printed low-voltage and high-performance OTFTs for circuit integration. In addition, some other interesting review papers [26–28] are recommended to readers to better understand gate dielectrics of OTFTs.

## 2. Mechanisms and requirements of dielectrics for printable OTFTs

OTFTs can also be called as organic field-effect transistors (OFETs), corresponding to the electric field that can control the behaviours of these devices. Since the demonstration of OTFTs in the mid 1980s [29], the device architecture and device physics have been widely studied and well developed. In this section, to better understand the dielectric function in printable OTFTs, the device architectures and the fundamental mechanisms and requirements of dielectrics are briefly introduced.