



# Polyimide/polyvinyl alcohol bilayer gate insulator for low-voltage organic thin-film transistors



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## ABSTRACT

In this paper, we report the fabrication of a polyimide/polyvinyl alcohol (PVA) bilayer gate insulator for low-voltage organic thin-film transistors (TFTs). The introduction of a PVA layer to form a bilayer structure improves the dielectric and insulating properties of the gate insulator. Organic TFTs with 150 nm-thick polyimide and PVA gate insulators were inactive at low operation voltages below 5 V. Conversely, organic TFTs with 150 nm-thick polyimide/PVA bilayer gate insulators exhibited excellent device performances. Our results suggest that the introduction of a PVA layer with a high dielectric constant could be a simple and efficient way to improve the device performance of low-voltage organic TFTs.

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## 1. Introduction

During the past decades, organic thin-film transistors (TFTs) have been extensively studied because of their potential application in low-cost and flexible electronics [1–10]. A variety of organic semiconductors for high-performance TFTs have been reported, while the development of organic gate insulators has been more limited. Recently, we reported the fabrication of polyimide gate insulators for high-performance organic TFTs [11–15]. Polyimide thin films have excellent electrical insulating properties. Polyimide gate insulators are not damaged by post wet processes such as semiconductor deposition and electrode patterning; furthermore, their surface properties can be modified by physical and chemical methods to improve the overall TFT performance [12–14]. It was reported that 2,7-didecyl[1]benzothieno[3,2-b][1]benzothiophene (C<sub>10</sub>-BTBT) TFTs with an alumina/polyimide gate insulator exhibited a field-effect mobility of 1.42 cm<sup>2</sup>/V s and an on/off current ratio of ~10<sup>6</sup> [14]. Quasi single-crystalline C<sub>10</sub>-BTBT TFTs with alkyl chain-grafted polyimide gate insulators exhibited a field-effect mobility of 1.74 cm<sup>2</sup>/V s and an on/off current ratio of ~10<sup>5</sup> [15]. Remarkably, organic TFTs with polyimide gate insulators showed excellent device performances. However, the dielectric properties of the polyimide gate insulators present some limitations. In general, the low-voltage operation of the TFTs is related to a high capacitance of the gate insulator layer;

for a high capacitance, small thickness or a high dielectric constant of the gate insulator is required. However, when decreasing the thickness of the polymeric gate insulator, the reduction of the electrical insulating and dielectric properties should be carefully considered. The density of the grain boundaries, defects, and pinholes could increase rapidly by decreasing the thickness of the polymeric gate insulator within a certain thickness range. Alternatively, the use of polymeric gate insulators with high dielectric constants has been proposed for low-voltage operation of organic TFTs [16–19]. The dielectric constants of the polyimide gate insulators are in the range of 2.8–3.4, which is considerably lower than that of the SiO<sub>2</sub> gate insulators (~3.9). For the low-voltage operation of organic TFTs, the dielectric constant of the polyimide gate insulators needs to be improved.

In this paper, we report on the polyimide/polyvinyl alcohol (PVA) bilayer gate insulators for low-voltage organic TFTs with a bottom-gate top-contact structure (Fig. 1). To overcome the relatively low dielectric constant of the polyimide gate insulator, a PVA layer with a high dielectric constant was introduced between the polyimide layer and the gate electrode. The dielectric constants of the 150 nm-thick polyimide gate insulator and the 150 nm-thick polyimide/PVA gate insulator were 2.7 and 6.4 at 100 Hz, respectively. At a low operation voltage below 5 V, pentacene and 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C<sub>8</sub>-BTBT) TFTs with the 150 nm-thick polyimide gate insulators were inactive, whereas pentacene and C<sub>8</sub>-BTBT TFTs with the 150 nm-thick polyimide/PVA gate insulators showed excellent device performances. The field-effect mobilities of the pentacene and C<sub>8</sub>-BTBT

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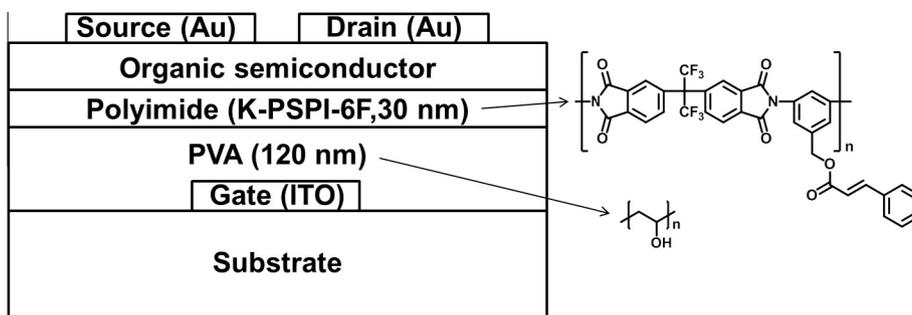


Fig. 1. Device structure of the organic TFT with the K-PSPI-6F/PVA bilayer gate insulator.

TFTs with the 150 nm-thick polyimide/PVA gate insulators were 0.20 and 0.88  $\text{cm}^2/\text{V s}$  at a low operation voltage below 5 V, respectively. We demonstrate that the introduction of the PVA layer could be a simple and efficient way to enhance the dielectric properties of the polyimide gate insulators and improve the device performance of low-voltage organic TFTs.

## 2. Experimental

For the synthesis of the polyimide (K-PSPI-6F), 0.01 mol of 4,4'-(hexafluoroisopropylidene)diphthalic anhydride (6FDA) and 0.01 mol of 3,5-diaminobenzyl cinnamate (DABC) were dissolved in 28.54 g of *m*-cresol. After the monomers were dissolved, 0.01 mol of isoquinoline was added and the solution was heated to 130 °C. When the viscosity of the reaction solution reached the saturation point, the polymerization was stopped. The polyimide was precipitated by adding the reaction mixture to the excess methanol. The precipitated solid was washed with methanol, filtered several times, and dried under vacuum. The purification process was repeated twice.

PVA ( $M_w$  146,000–186,000, 99 + % hydrolyzed) was dissolved in water (3 wt%), and spin-coated on patterned indium tin oxide (ITO)/glass substrates. The spin-coated films were annealed at 100 °C for 60 min. The thickness of the PVA layers was controlled to be 120 nm. K-PSPI-6F was dissolved in cyclohexanone (1 wt%) and spin-coated on the PVA layer. The spin-coated films were annealed at 90 °C for 2 min and exposed to ultraviolet (UV) light at a dose of 3 J/cm<sup>2</sup>. Then, the films were annealed at 130 °C for 30 min. The final thickness of the K-PSPI-6F/PVA films was controlled to be 150 nm.

For the measurement of the capacitance and leakage current density, metal–insulator–metal (MIM) devices were prepared by deposition of a top gold electrode on the gate insulator-coated ITO/glass substrate. The active area of the MIM devices was 50.24 mm<sup>2</sup>. For the electrical characterization, bottom-gate top-contact organic TFT devices were prepared. To produce the gate electrode, ITO was patterned (2 mm wide stripes). A 60 nm-thick organic semiconductor was deposited onto the gate insulator layer through a shadow mask by using thermal evaporation at a pressure of  $3 \times 10^{-6}$  Torr. The evaporation rate of the organic semiconductor was 0.3 Å/s. The organic TFTs were then completed by deposition of 50 nm-thick source and drain gold electrodes onto the semiconductor layer through a shadow mask by using thermal evaporation, creating transistors with a channel length (*L*) and a width (*W*) of 50 and 1000  $\mu\text{m}$ , respectively.

The thicknesses of the gate insulator layers were determined with an alpha-step surface profiler (a-step DC50, KLA-Tencor). Atomic force microscopy (AFM) images were obtained with a tapping mode microscope (Nanoscope IV, Digital Instrument). The capacitance was measured with an Agilent 4294A impedance

analyzer. The output and transfer characteristic curves of the organic TFTs were measured with an Agilent E5272 semiconductor parameter analyzer. All these electrical measurements were performed in air without any encapsulation.

## 3. Results and discussion

The K-PSPI-6F is a photo-sensitive soluble polyimide (Fig. 1). Polymers with cinnamate groups could be crosslinked by the UV exposure [20–23]. Although the crosslinked 300 nm-thick K-PSPI-6F films exhibited excellent electrical insulating property [13], the dielectric properties have some limitations. Due to the low dielectric constant,  $\sim 2.8$ , the 300 nm-thick K-PSPI-6F gate insulator is not suitable for low-voltage operation of the organic TFTs, as, to this end a small thickness or a high dielectric constant of the gate insulator is required. To increase the capacitance of the K-PSPI-6F gate insulator, we prepared 150 nm-thick K-PSPI-6F films. Fig. 2a shows the capacitance of the 150 nm-thick K-PSPI-6F film as a function of the frequency. The capacitance and dielectric constant of the 150 nm-thick K-PSPI-6F film at 100 Hz were measured to be 160 pF/mm<sup>2</sup> and 2.7, respectively. Fig. 2b shows the leakage current density of the 150 nm-thick K-PSPI-6F film as a function of the electric field. The leakage current density of the 150 nm-thick K-PSPI-6F film at 2 MV/cm was measured to be  $4.8 \times 10^{-5}$  A/cm<sup>2</sup>, which is considerably higher than that of the 300 nm-thick K-PSPI-6F film ( $1.6 \times 10^{-10}$  A/cm<sup>2</sup>) [13]. The electrical insulating properties of the polymer films could decrease rapidly by decreasing the film thickness within a certain thickness range because of the undensified state of the thin film. We prepared pentacene and C<sub>8</sub>-BTBT TFTs with a 150 nm-thick K-PSPI-6F gate insulators, and observed that they were inactive at low operation voltages below 5 V.

The capacitance of the gate insulator might need to be further increased for low-voltage operation. To overcome the relatively low dielectric constant of the polyimide, we prepared K-PSPI-6F/PVA bilayer gate insulators. Although PVA has a high dielectric constant, its use as a gate insulator is limited because of the massive hydroxyl groups, which are related to the large current hysteresis and high off current of the organic TFTs [24–29]. The K-PSPI-6F/PVA bilayer gate insulator could exhibit complementary properties. To obtain the bilayer gate insulator, a 30 nm-thick K-PSPI-6F layer was deposited on a 120 nm-thick PVA layer. The capacitance and dielectric constant of the bilayer film at 100 Hz were measured to be 378 pF/mm<sup>2</sup> and 6.4, respectively (Fig. 3a). For comparison, we also prepared MIM devices with PVA layers. The capacitance and dielectric constant of the 150 nm-thick PVA film at 100 Hz were measured to be 426 pF/mm<sup>2</sup> and 7.2, respectively (Fig. 3a). Owing to the small thickness of the K-PSPI-6F layer, the capacitance and dielectric constant of the 150 nm-thick bilayer film decreased only slightly

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