

Effects of interfacial tension and molecular dipole moment on the electrical characteristics of low-voltage-driven organic electronic devices

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ABSTRACT

In organic electronic/photonics devices, numerous types of interfaces and their properties exhibit profound correlation with device performance. For optimizing the performance of organic electronic/photonics devices, appropriate and effective interface engineering needs to be developed. In this study, a high dielectric constant material, hafnium dioxide (HfO_2), and an organic semiconductor, *N,N'*-ditridecyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI- C_{13}) were adopted as the dielectric and active layers, respectively, to fabricate low-voltage-driven organic thin-film transistors. Three kinds of insulating polymers were selected to serve as buffer layers (BLs) to modify HfO_2 . After the addition of BLs onto HfO_2 , the insulating properties of HfO_2 and the microstructures of PTCDI- C_{13} active layers improved, resulting in considerably enhanced electrical characteristics and stability of the devices. Among different polymeric BLs, the BL polymer exhibiting smaller interfacial tension with PTCDI- C_{13} can induce PTCDI- C_{13} to form better microstructures and generate lower interfacial trap density despite the rougher topography of polymeric BL, leading to improved electrical characteristics of the corresponding device. However, we observed that BL polymer with larger dipole moment of side groups can yield better electrical stability of the corresponding device under continuous operation compared with polymers with smaller interfacial tension. During long-term operation, the dipoles can be aligned by an electric field and form a strong dipole layer to facilitate charge accumulation and alleviate device degradation caused by bias-stress-induced trap/defect states. We further adopted a BL polymer with both small interfacial tension and large dipole moment to fabricate low-voltage-driven organic complementary inverters. The inverter can exhibit high electrical characteristics and stability during continuous operation. Interfacial tension and molecular dipole moment are possible important issues for effective interface engineering.

1. Introduction

Organic semiconductors have attracted considerable attention owing to their high potential for electronic/photonics products with low cost, large area, light weight, and flexibility. In the circuits of various electronic/photonics products, switch and logic devices, such as complementary inverters, are essential components composed of transistors. Hence, organic thin-film transistors (OTFTs) constitute an important application of organic semiconductors. The electrical performance of OTFTs is strongly correlated with the microstructural and dielectric properties of active and dielectric layers, respectively [1,2]. The microstructural properties of active layers influence the behavior of charge transport and charge mobility (μ) of OTFT devices [2–4]. Good microstructures of active layers cause efficient charge transport, leading to high μ and drain current (I_D) of OTFTs. The dielectric properties of

dielectric layers affect charge accumulation in the conduction channels and the leakage current of OTFT devices [5–7]. Good dielectric layers facilitate charge accumulation to form conduction channels and suppress leakage current, resulting in low threshold voltage (V_T) and high gate control capability of OTFTs. For commercialized applications of OTFT devices, such as flat panel displays and wearable electronic devices, low-voltage-driven OTFTs are required. Dielectric layers must possess high capacitance and facilitate the formation of conduction channels at low voltage to achieve low-voltage-driven OTFTs [5,8,9].

Various kinds of dielectric layers, including high-dielectric-constant (high- κ) materials, crosslinked polymers, self-assembled materials, electrolyte/ion-gel, and interfacial dipole layers, have been proposed to obtain high capacitance. High- κ materials, such as metal oxides (MOs) [10–12] and insulating polymers with strong polar groups [13,14], can generate strong polarization field to achieve high capacitance.

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Crosslinked insulating polymers [8,15,16] and self-assembled insulating molecules [9,17,18] can form densely packed structures at low thickness to maintain good insulating properties and produce high capacitance. The ions of electrolyte/ion-gel under an electric field can be aligned, create strong polarization field, and result in high capacitance [19,20]. Interfacial dipole layers originating from the bonding of insulating polymer with MO can be substantially enhanced under an electric field to generate strong dipole field and achieve high capacitance [21].

Among a variety of techniques for obtaining high-capacitance dielectric layers, the adoption of high- κ MOs is a simple, effective, and commonly used method [5,12,22]. However, the MO surface tends to be rough and generates numerous trap/defect states. When MOs are applied as dielectric layers, the trap/defect states existing at the interface between active layers and MOs can hinder charge transport and reduce the performance of OTFTs [22–24]. In addition, organic semiconductors grown on MOs often evolve into poor microstructures because of the rough surface and the mismatch of surface energy between organic semiconductors and MOs, leading to inefficient charge transport and poor performance of OTFTs [25–27]. The interfacial trap/defect states and poor microstructures of active layers also tend to degrade the electrical stability of OTFTs during long-term operation [22,24]. Therefore, effective interface engineering to modify the interface of MOs with active layers is needed for the performance and stability optimization of low-voltage-driven OTFTs.

In this study, we adopt the high- κ material hafnium dioxide (HfO_2) and the organic semiconductor *N,N'*-ditridecyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI- C_{13}) as dielectric and active layers, respectively, to fabricate low-voltage-driven OTFT devices. Three types of insulating polymers, including poly (methyl methacrylate) (PMMA), crosslinked poly (4-vinylphenol) (CPVP), and polyimide (PI), are selected as buffer layers (BLs) to modify the HfO_2 surface. After polymeric BLs are deposited on HfO_2 , the electrical performance and stability of PTCDI- C_{13} -based OTFTs are enhanced, resulting from simultaneous improvement in the insulating properties of dielectric layers, microstructures of PTCDI- C_{13} , and interfacial properties of PTCDI- C_{13} with HfO_2 /BLs. Moreover, PTCDI- C_{13} -based OTFTs with different polymeric BLs exhibit contrasting electrical characteristics and stability. We observe that instead of a smooth HfO_2 /BL surface, the small interfacial tension between polymeric BL and PTCDI- C_{13} can cause low interfacial trap density and superior microstructures of PTCDI- C_{13} , resulting in improved electrical characteristics of devices. However, the strong dipole moment of side groups of the polymeric BL can facilitate charge accumulation to increase the electrical stability of devices during long-term operation in addition to PTCDI- C_{13} microstructures. We also applied an insulating polymer with both small interfacial tension and strong dipole moment of side groups as the BL to fabricate low-voltage-driven organic complementary inverters. The inverters exhibit high electrical characteristics, such as high signal gain, high logic reliability, low power consumption, and high electrical stability under continuous operation.

2. Experimental section

2.1. Device preparation

The device structure of PTCDI- C_{13} -based OTFTs used in this study is illustrated in Fig. 1a. A cleaned indium-tin-oxide (ITO) glass substrate was used as a gate electrode. A 100 nm-thick HfO_2 film as a dielectric layer was grown on ITO by using a radio frequency sputter system at a power of 120 W with an argon/oxygen gas mixture of 40/20 sccm. Subsequently, the HfO_2 thin film was annealed at 200 °C for 1 h. Various insulating polymeric materials, including PMMA, CPVP, and PI, were spin coated onto HfO_2 as BLs. Details on the preparation and processing of polymeric BLs are described in the supplementary material. After the fabrication of polymeric BLs, the 60 nm-thick PTCDI- C_{13}

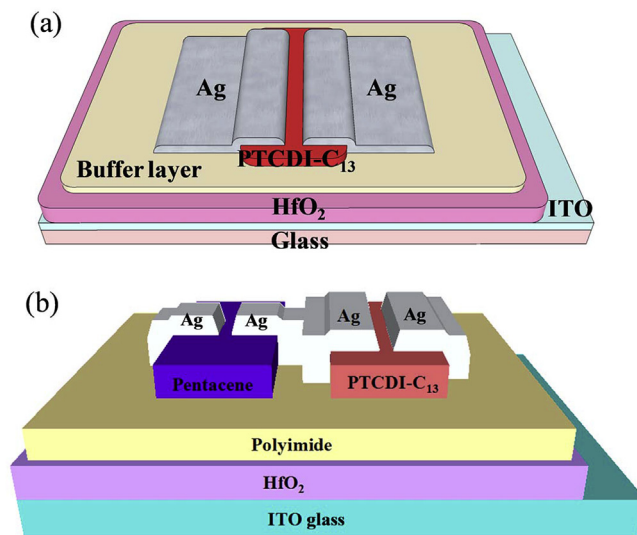


Fig. 1. Device structures of (a) PTCDI- C_{13} -based OTFT and (b) organic complementary inverter.

active layer, which was synthesized in our laboratory, was thermally evaporated on polymeric BLs at a deposition rate of 0.2 Å/s. Finally, 70 nm silver (Ag) was thermally evaporated on PTCDI- C_{13} at a deposition rate of 0.4 Å/s to form the source and drain electrodes through a shadow mask. The channel width (W) and length (L) of OTFT devices were 2000 and 100 μm , respectively. For fabricating the organic complementary inverters, the processes of the ITO gate electrode, HfO_2 dielectric layer, and PI BL involved the same procedures described for the above OTFT devices. After the PI BL was obtained, 60 nm pentacene (purity: 98%, Sigma-Aldrich Co.) and PTCDI- C_{13} were thermally evaporated at a deposition rate of 0.2 Å/s on the PI BL through shadow masks to act as p -type and n -type active layers, respectively. Subsequently, the Ag source and drain electrodes were deposited on pentacene and PTCDI- C_{13} to complete the organic complementary inverter (Fig. 1b). The W values of p -type and n -type channels were 500 and 1500 μm , respectively, and both L values were 100 μm .

2.2. Sample characterization

We used a metal/insulator/metal (MIM) structure to study the dielectric properties of dielectric films. Bare HfO_2 and HfO_2 /BL thin films were deposited on ITO glass substrates. Ag was thermally evaporated on both films to complete the MIM structures. The current-voltage and capacitance-voltage curves of dielectric films and the electrical characteristics of OTFTs and organic complementary inverters were measured using a Keithley 4200SCS semiconductor parameter analyzer. All electrical measurements were performed in a nitrogen-filled glove box. The surface morphologies of thin films were obtained via Park XE-100 atomic force microscopy. The microstructures of PTCDI- C_{13} thin films were analyzed using a Rigaku RINT 2000 X-ray diffractometer with an X-ray wavelength of 1.5406 Å under a scan step of 0.01°. The surface free energies of all films, including the semiconductor and dielectric films, were obtained from the DataPhysics OCA contact angle system.

3. Results and discussion

3.1. Dielectric layers

Fig. 2 shows the dielectric properties of HfO_2 thin films capped with different BLs. The capacitance of the bare HfO_2 thin film is higher than 80 nF/cm² and maintains a constant value in the voltage sweep region as shown in Fig. 2a. After spin coating of various polymeric BLs onto HfO_2 , the equivalent capacitances of all specimens exhibit significant

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