



High performance nitrogen dioxide sensor based on organic field-effect transistor utilizing ultrathin CuPc/PTCDI-C8 heterojunction



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ABSTRACT

Organic field-effect transistors (OFETs) with a heterojunction structure, consisting of the p-type and n-type organic semiconducting materials of copper phthalocyanine (CuPc) and the dioctyl perylene tetracarboxylic diimide (PTCDI-C8), respectively, were fabricated. The heterojunction OFETs were used as the nitrogen dioxide (NO₂) gas sensors, and the sensing properties were characterized with the variation of PTCDI-C8 layer thicknesses. The results showed that the OFET sensors with the optimized film thickness of 0.5 nm PTCDI-C8 and 7 nm CuPc, had one order of magnitude enhancement of sensitivity, compared to the device with single CuPc active layer. The sensitivity improvement was attributed to the intensification of the charge transfer in the heterojunction structure while introducing the oxidizing gas of NO₂.

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1. Introduction

Organic field-effect transistors (OFETs) have received lots of attention due to their wide range of electronic applications, such as flexible electronic papers, flat-panel displays, radio frequency identification (RFID) tags, and logic circuits [1–7]. In view of their merits of low cost, low power consumption, room-temperature operating condition, as well as the signal amplifying property, the OFETs are considered to be promising candidates for chemical-, physical-, and bio-sensors [8–11]. As widely reported in literatures, the sensing mechanism of OFET-based gas sensors can be summarized as the electrical characteristics changes, including the field-effect mobility, current on/off ratio, threshold voltage and the channel current in OFETs, induced by the interaction between the analyte molecules and the organic semiconducting layers [12]. The sensing signal can be direct readout from the electrical performance variation.

For the past decades, a lot of efforts have been made to achieve superior performance of OFET-based sensors, such as reducing the channel length, enhancing the interaction between analyte and the functional layer, using OFET-based circuits, and employing heterojunction structure as sensing active layer [13–17]. Among

these methods, OFETs with heterojunction structure active layer have been proven to be an effective route to enhance sensing properties for OFETs [18,19]. In heterojunction OFET devices, the combination of two or more functional semiconducting materials with their properties, can not only maintains the intrinsic property of each material but also extends the device function [20,21].

As one of the most dangerous combustion exhaust pollutions, nitrogen dioxide (NO₂) has attracted extensive focus. While OFET sensor based on heterojunction structure is emerging as an efficient detection method for NO₂ gas [22,23]. Recently, Ji et al. reported an OFET based on highly ordered ultra-thin heterojunction organic film, enabling room-temperature detection of NO₂ [24]. They introduced *N,N'*-diphenyl perylene tetracarboxylic diimide (PTCDI-Ph) film on top of para-hexaphenyl (p-6P) film. The resulted sensor devices exhibit greater sensitivity to NO₂ gas than that of single active layer p-6P OFET sensors. Such results indicated the potential of heterojunction OFET with PTCDI and their derivatives as one of the functional layer for enhancing NO₂ sensing. Compared to PTCDI-Ph, PTCDI-C8 has good solubility and higher mobility [25,26]. Besides, CuPc based OFETs has been used widely in NO₂ sensing area due to its stability and high sensing performance [27]. However, heterojunction of CuPc and PTCDI-C8 has not been reported. And the sensing properties to chemical gases of OFET sensors are highly related to the thickness of OFET active layer. Thus, to realize high performance NO₂ sensors,

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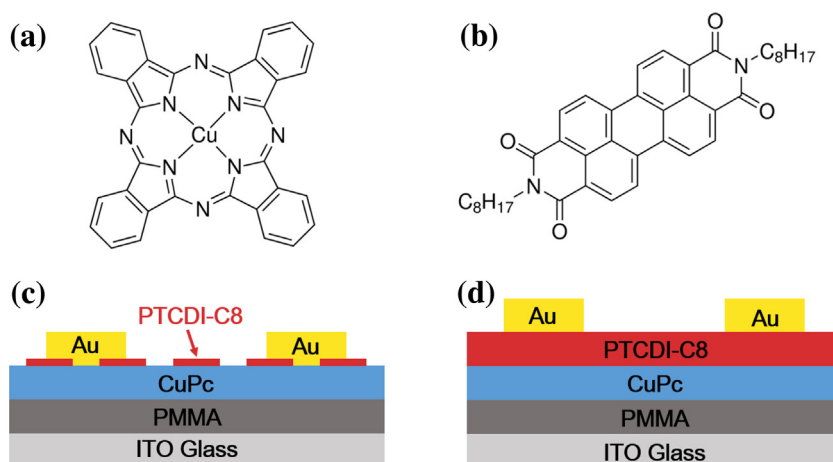


Fig. 1. Molecular structures of (a) CuPc and (b) PTCDI-C8. Device architectures of the heterojunction OFET sensor with (c) discontinuous and (d) continuous PTCDI-C8 layers.

it is very important to understand the heterojunction film thickness influence on OFET sensor.

To systemically investigate the relationship between sensing properties and thicknesses of each layer in heterojunction, in this work, the heterojunction OFET-based sensors with various thickness of n-type PTCDI-C8 on p-type copper phthalocyanine (CuPc) were fabricated, and the NO₂ sensing performance of these OFET-based sensors was characterized. With the optimized PTCDI-C8 film thickness, the heterojunction OFET-based sensors exhibit a significant increase in NO₂ gas sensitivity under room temperature, enabling a detection limitation of NO₂ as low as 2 ppm. This study provides a promising prospect for the application of OFET in room temperature, low-cost and high sensitivity NO₂ gas detectors, and describes the importance of active layer thickness to optimize the sensing properties of OFETs.

2. Experimental

The molecular structures of PTCDI-C8 and CuPc are shown in Fig. 1a and b, and the bottom gate top-contacted configurations of OFETs with discontinuous and continuous PTCDI-C8 are shown in Fig. 1c and d, respectively. The indium tin oxide (ITO) coated glass was used as substrates and gate electrodes, respectively. Firstly, the ITO glass substrates were successively ultrasonic cleaned in detergent, acetone, deionized water and isopropyl alcohol for 15 min each. Then, a 520 nm thick poly(methyl methacrylate) (PMMA) dielectric layer was spin-coated on the substrates, and baked at 150 °C for 1 h in air to remove the solvent residue. Thirdly, 7 nm CuPc film and ultrathin PTCDI-C8 layer with thickness ranging from 0 to 2 nm were deposited, respectively, under 3×10^{-4} Pa at a rate of 0.5 Å/s. Finally, a 50 nm thick gold (Au) was

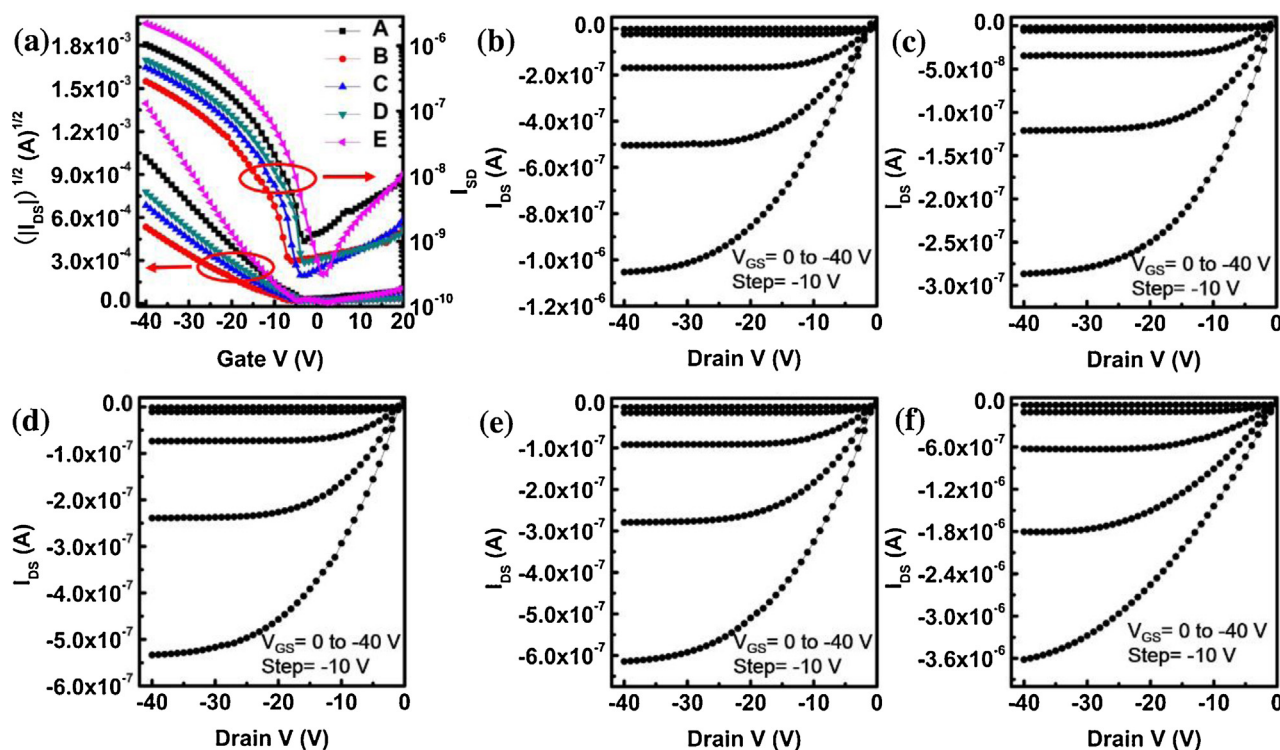


Fig. 2. (a) Transfer curves of the samples A–E. Output curves of (b) sample A, (c) sample B, (d) sample C, (e) sample D, and (f) sample E.

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