



# Materials and interfaces issues in pentacene/PTCDI-C<sub>8</sub> ambipolar organic field-effect transistors with solution-based gelatin dielectric

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## ABSTRACT

Gelatin is a natural protein, which works well as the gate dielectric for pentacene/N,N-di-*o*-octyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI-C<sub>8</sub>) ambipolar organic field-effect transistors (OFETs) in air ambient and in vacuum. An aqueous solution process was used to form the gelatin gate dielectric film on poly(ethylene terephthalate) (PET) by spin-coating and subsequent casting. Pentacene morphology and interface roughness are two major factors affecting the electron and hole field-effect mobility ( $\mu_{FE}$ ) values of pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs in vacuum and in air ambient. In contrast, water absorption in gelatin has higher contribution to the electron and hole  $\mu_{FE}$  values in air ambient. The ambipolar performance of pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs depends on their layer sequence. For example, when PTCDI-C<sub>8</sub> is deposited onto pentacene, i.e. in the structure of PTCDI-C<sub>8</sub>/pentacene, unbalanced ambipolar characteristics appear. In contrast, better ambipolar performance occurs in the structure of pentacene/PTCDI-C<sub>8</sub>. The optimum ambipolar characteristics with electron  $\mu_{FE}$  of 0.85 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and hole  $\mu_{FE}$  of 0.95 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> occurs at the condition of pentacene (40 nm)/PTCDI-C<sub>8</sub> (40 nm). Surprisingly, water absorption plays a crucial role in ambipolar performance. The device performance changes tremendously in pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs due to the removal of water out of gelatin in vacuum. The optimum ambipolar characteristics with electron  $\mu_{FE}$  of 0.008 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and hole  $\mu_{FE}$  of 0.007 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> occurs at the condition of pentacene (65 nm)/PTCDI-C<sub>8</sub> (40 nm). The roles of layer sequence, relative layer thickness, and water absorption are proposed to explain the ambipolar performance.

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## 1. Introduction

Ambipolar organic field-effect transistors (OFETs) may exhibit either p-channel or n-channel characteristics, which are attractive in the fields of organic complementary metal oxide semiconductor (CMOS) circuits [1–4]. Three kinds of structural designs of organic semiconductor such as bilayer, blend and single-component have been

developed for ambipolar OFETs [5–9]. The bilayer structure has been the most common one, which is formed by depositing p- and n-channel semiconductor in sequence on the gate dielectric in ambipolar OFETs. The performance of ambipolar OFETs with the bilayer structure depends mainly on the pair of organic semiconductors, relative layer thickness and source/drain electrodes [10,11]. For instance, several pairs of organic semiconductors such as CuPc/F<sub>16</sub>CuPc, pentacene/PTCDI-C<sub>8</sub>, BP2T/F<sub>16</sub>CuPc, pentacene/PTCDI-C<sub>13</sub>H<sub>27</sub> and C<sub>60</sub>/pentacene exhibit comparable  $\mu_{FE}$  of hole and electron leading to very good ambipolar

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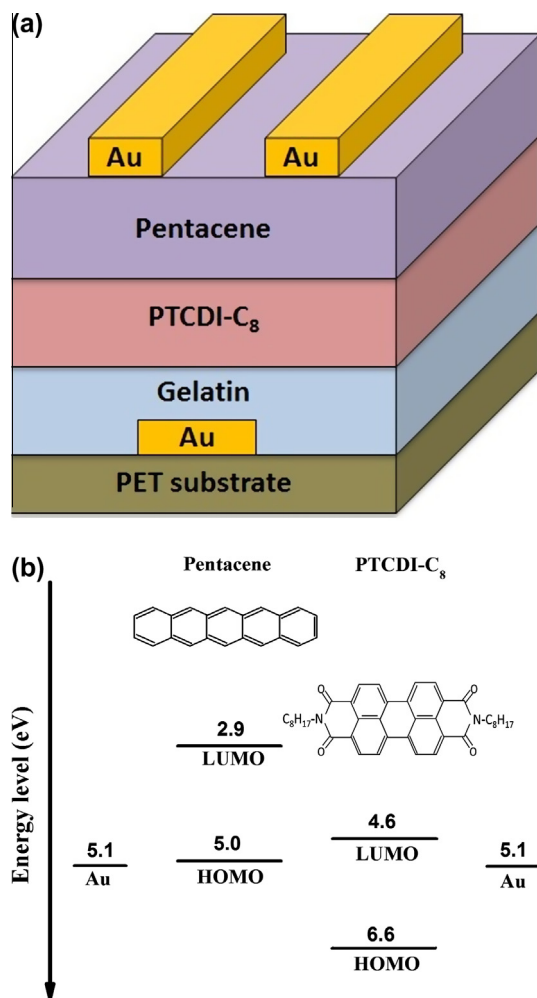
transport behavior [10–17]. However, the ambipolar characteristics degrade when the bilayer thickness deviates from the optimum condition, e.g. CuPc (10 nm)/F<sub>16</sub>CuPc (2 nm) in the CuPc/F<sub>16</sub>CuPc ambipolar OFETs reported by Wang et al. [10]. Moreover, asymmetric Au and Mg as source/drain electrodes may enhance hole and electron injection in the PTCDI-C<sub>13</sub>H<sub>27</sub>/pentacene ambipolar OFETs, reported by Rost et al. [11]. The ambipolar device exhibit electron and hole  $\mu_{FE}$  of  $3 \times 10^{-3}$  and  $1 \times 10^{-4}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, respectively.

Most bilayer ambipolar OFETs studied to date exhibit low  $\mu_{FE}$  from  $1 \times 10^{-4}$  to 0.2 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and high threshold voltage ( $V_{TH}$ ) from 7 to 66 V, which limit further practical applications [10–17]. Note that  $\mu_{FE}$  and  $V_{TH}$  are closely correlated to the pair of organic semiconductors. For example, hole  $\mu_{FE}$  of 0.2 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> with  $V_{TH}$  of -2.3 V and electron  $\mu_{FE}$  of 0.04 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> with  $V_{TH}$  of 66 V was reported for C<sub>60</sub>/pentacene ambipolar OFETs by Yan et al. [17]. In principle, the drawbacks of low  $\mu_{FE}$  and high  $V_{TH}$  may be improved if organic ambipolar OFETs are constructed with organic semiconductors with high  $\mu_{FE}$  and low  $V_{TH}$ . Note that the performance of OFETs also influenced strongly by the gate dielectric. Recently, a p-channel pentacene OFET with high hole  $\mu_{FE}$  of 16 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and low  $V_{TH}$  of -1 V has been illustrated by using gelatin as the gate dielectric reported by Mao et al. [18]. A n-channel PTCDI-C<sub>8</sub> OFET with high electron  $\mu_{FE}$  of 0.74 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and low  $V_{TH}$  of 2.6 V has also been presented [19]. These may open the possibility of using gelatin as the gate dielectric to fabricate ambipolar OFETs with high  $\mu_{FE}$  and low  $V_{TH}$ .

In this article, we present the device performance of pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs with gelatin as the gate dielectric. Although pentacene and PTCDI-C<sub>8</sub> exhibit different molecular shape, crystal packing and grain size, PTCDI-C<sub>8</sub> is chosen to be a counterpart of pentacene to form a bilayer in ambipolar OFETs based on two reasons. First, the device characteristics of pentacene or PTCDI-C<sub>8</sub> OFETs with SiO<sub>2</sub> as the gate dielectric are well characterized and can be used for comparison. Second, the pentacene/PTCDI-C<sub>8</sub> bilayer was reported to exhibit ambipolar characteristics [12]. The role of gelatin in the device performance of pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs can be understood by comparing with previously published data.

## 2. Materials and methods

The bottom gate configuration was chosen to fabricate pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs with gelatin as the gate dielectric, which is schematically shown in Fig. 1a. The preparation of an aqueous solution of the type A gelatin of 300 bloom can be found in our previously published paper [18]. A gelatin thin film was coated onto a poly(ethylene terephthalate) (PET) patterned with 70 nm Au gate electrodes by spin-coating and subsequent casting at 60 °C for 36 h. In order to reduce the gate leakage current in pentacene/PTCDI-C<sub>8</sub> ambipolar OFETs, the film thickness of gelatin was adjusted by the feed-back of device data. PTCDI-C<sub>8</sub> (99%, Sigma–Aldrich) was then thermally evaporated as the n-channel semiconductor onto the cast gelatin thin film at room temperature (RT) through a shadow



**Fig. 1.** (a) Schematic of a pentacene/PTCDI-C<sub>8</sub> ambipolar OFET with gelatin as the gate dielectric and (b) work function of the Au metal and energy levels (HOMO and LUMO) of pentacene and PTCDI-C<sub>8</sub>. The chemical structures of pentacene and PTCDI-C<sub>8</sub> molecules are inserted in the figure.

metal mask at a base pressure of  $1 \times 10^{-6}$  torr. Pentacene (99%, Sigma–Aldrich) of various thicknesses was subsequently deposited at RT onto the PTCDI-C<sub>8</sub> layer to form the bilayer structure. The deposition rates of PTCDI-C<sub>8</sub> and pentacene were kept at 0.3 Å/s monitored by a quartz crystal oscillator. The morphology of the PTCDI-C<sub>8</sub> and pentacene layer was measured using atomic force microscope (AFM). A 70-nm-thick Au was finally thermally deposited onto pentacene/PTCDI-C<sub>8</sub> to define source and drain electrodes. The channel length and width were 50 μm and 600 μm, respectively. The output and transfer curves of ambipolar OFETs were measured using Agilent 4155C. Quasi-static capacitance–voltage (C–V) curves were taken by sweeping voltage across the Au/gelatin/Au structure using Agilent B1500. The sweeping rate of voltage in the quasi-static capacitance measurements was kept the same as that used in the transfer curve measurements. The quasi-static capacitance value was more accurate in the derivation of  $\mu_{FE}$  [18].

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