



A study of effects of electrode contacts on performance of organic-based light-emitting field-effect transistors

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ABSTRACT

Herein is presented a comparative performance analysis of heterojunction organic-based light-emitting field-effect transistors (OLEFETs) with symmetric (Au only) and asymmetric (Au and LiF/Al) electrode contacts. The devices had a top source–drain contact with long-channel geometry and were produced by sequentially depositing *p*-type pentacene and *n*-type *N,N'*-ditridecylperylene-3,4,9,10-tetracarboxylic diimide (P13) using a neutral cluster beam deposition apparatus. The spectroscopic, structural and morphological properties of the organic thin films were examined using photoluminescence (PL) spectroscopy, X-ray diffraction (XRD) method, laser scanning confocal and atomic force microscopy (LSCM, AFM). Based upon the growth of high-quality, well-packed crystalline thin films, the devices demonstrated ambipolar field-effect characteristics, stress-free operational stability, and light emission under ambient conditions. Various device parameters were derived from the fits of the observed characteristics. The hole mobilities were nearly equal irrespective of the electrode contacts, whereas the electron mobilities of the transistors with LiF/Al drain electrodes were higher due to the low injection barrier. For the OLEFETs with symmetric electrodes, electroluminescence (EL) occurred only in the vicinity of the hole-injecting electrode, whereas for the OLEFETs with asymmetric electrodes, the emission occurred in the vicinity of both hole- and electron-injecting electrodes. By tuning the carrier injection and transport through high- and low-work function metals, the hole-electron recombination sites could be controlled. The operating conduction and light emission mechanism are discussed with the aid of EL images obtained using a charge-coupled device (CCD) camera.

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1. Introduction

In manufacturing organic-based integrated circuits (ICs), simplifying the circuit and reducing the number of components are crucial [1–6]. In that sense, the combined functionalities of both the electrical switching of organic field-effect transistors (OFETs) and the luminescence of organic light-emitting diodes (OLEDs) in an optoelectronic device known as an organic light-emitting field-effect transistor (OLEFET) provides a new option in designing complex functional organic devices. An electrical switching operation is achieved by modulating the current between the source and drain electrodes by tuning the gate voltage, while electroluminescence (EL) occurs by hole-electron recombination in the active channel [7–10]. Since the fabrication of π -conjugated tetracene-based OLEFET was first reported in 2003, considerable technological

progress has been made [1]. The OLEFET array is one of the simplest IC devices available and increases the range of prospective applications of organic-based optoelectronic devices such as flat panel displays, optical communication devices, and electrically driven organic lasers [7–14].

Balanced hole-electron carrier concentrations and formation of the singlet exciton within the active channel are essential in producing high-performance OLEFETs [9]. Most OLEFETs use single organic and polymeric compounds showing either *p*- or *n*-type unipolar behaviors; the majority of carriers are either holes or electrons. As a result, typical unipolar OLEFETs, including the early tetracene-based devices, suffer an unavoidable hole-electron imbalance and strong exciton quenching at the metal electrode. Carrier balance can be realized by employing single ambipolar materials [15,16] or by combining two unipolar materials through blended [17] or bi-layered structures [13,14,18]. In most ambipolar single-component- and blend-based OLEFETs, however, efficient EL was not attained due to the difficulties in achieving a balanced

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injection and carrier transport. In contrast, despite achieving physical separation and growth incompatibility between the *p*- and *n*-type layers, the bi-layered, heterojunction-based OLEFETs with high carrier mobilities showed good balanced ambipolarity and EL efficiency. Another scheme to avoid carrier imbalance is to use several metal electrodes with different work functions (WFs) [19]. Typical high-WF metals like Au (5.1 eV) are suitable for hole injection into the highest occupied molecular orbitals (HOMO) of the *p*-type organic layer, while low-WF metals such as Ca (2.8 eV) [20], Mg (3.7 eV) [21], LiF/Al (2.9 eV) [22,23], etc. are suitable for electron flowing into the lowest unoccupied molecular orbitals (LUMO) of the *n*-type organic layer. By adopting an asymmetric electrode configuration, more facile charge-carrier injection and balanced carrier concentrations can be achieved.

The preparation of crystalline organic layers is another essential aspect in the manufacturing of high-performance OLEFETs. Unlike common vapor deposition and/or solution-processing methods, the less popular method using neutral cluster beam deposition (NCBD) in this work is simple, but it is promising for producing various thin-film devices. Cluster beams have been widely used in gas-phase kinetics and dynamics to examine the effects of solvation and intermolecular interactions at the molecular level. As the vapor-phase molecules undergo adiabatic supersonic expansion in a high vacuum, neutral cluster beams of weakly bound organic molecules are generated at the throat of the nozzle. The unique characteristics of a neutral cluster beam are its high translational kinetic energy and directionality, which allow the precise formation of smooth, uniform films after collision with the target substrate. The novel NCBD scheme shows significant improvements in the surface morphology, crystallinity, packing density, and room-temperature substrate deposition, the distinctive advantages of which cannot be achieved easily through traditional vapor deposition techniques. A series of NCBD-based optoelectronic devices were recently produced and successfully characterized [2,3,13,14,24–31].

In this paper, we present the fabrication and comparative characterization of air-stable, ambipolar heterojunction-based OLEFETs with symmetric (Au only) and asymmetric (Au and LiF/Al) electrode contacts. Devices with a top source–drain contact and long-channel geometry were produced by sequentially depositing *p*-type pentacene and *n*-type *N,N'*-ditridecylperylene-3,4,9,10-tetracarboxylic diimide (P13) as hole- and electron-transporting materials, respectively, using the NCBD method. The effect of the electrode contacts on the OLEFET performance was examined by tuning the electron injection and transport through high WF Au (5.1 eV) vs. low WF LiF/Al (2.9 eV) metals. Here, unlike most low-WF metals that are unstable in air, by inserting an ultrathin LiF layer (<1 nm thick) between an Al electrode and the P13 organic layer, the electron injection, air-stability and EL efficiency could be improved [23]. Fig. 1 shows the molecular structures and positions of HOMOs and the lowest unoccupied molecular orbitals (LUMO) of pentacene and P13. The relative locations of energy levels are well-matched to produce singlet excitons for efficient EL. We initially focused on characterization of the pentacene and P13 active layers using photoluminescence (PL) spectroscopy, X-ray diffraction (XRD) method, laser scanning confocal and atomic force microscopy (LSCM, AFM). The device parameters of the heterojunction-based OLEFETs such as the hole and electron carrier mobilities and EL are then deduced from the observed characteristics such as current-voltage (*I*-*V*) and current-voltage-light emission (*I*-*V*-*L*) plots. The devices described herein showed good ambipolar characteristics, stress-free operational stability, and EL under ambient conditions. The operating conduction and light emission mechanism accounting for the observed EL are discussed with the aid of

EL camera images.

2. Experimental

Fig. 1 shows schematic views of the OLEFET devices with the molecular structures of pentacene and P13 (Sigma-Aldrich), and the corresponding energy-level diagram. Two types of top-contact OLEFETs with symmetric and asymmetric electrode contacts were produced. The substrates were made up of a heavily *n*-doped Si substrate as a common gate electrode and 2000 Å-thick SiO₂ thermally grown atop the substrates as a gate dielectric. A series of successive ultrasonic treatments were utilized to improve the device performance using acetone, 20% HNO₃, hot trichloroethylene, methanol, and deionized water, followed by blowing dry with N₂ gas. Then, the substrates were exposed to 254-nm UV radiation for 15 min.

The authors' laboratory-made NCBD system was utilized to prepare the organic active layers. The apparatus was explained in detail elsewhere [24], and only a relevant account is presented here. The NCBD vacuum chamber consisted of a pair of evaporation crucibles, a drift region, and a substrate. The pentacene sample placed in an evaporation crucible was first heated resistively between 500 and 520 K and a highly directional, weakly bound pentacene cluster beam was subsequently formed at the throat of the nozzle. The beam experienced adiabatic supersonic expansion into the drift zone at a working pressure of approximately 6×10^{-6} Torr and was directly deposited onto the substrates. P13 placed in a separate evaporation crucible was heated between 480 and 510 K and deposited on top of the pentacene layer. The optimized thickness and deposition rate were 40 Å at 0.7–1.0 Å/s for pentacene, and 350 Å at 0.7–1.0 Å/s for P13.

A comparative study was executed to inspect the effect of electrode contacts on OLEFET performance. For the OLEFETs with symmetric Au source and drain electrodes, Au was thermally evaporated onto the P13 layer through a shadow mask deposited at a rate of 2–4 Å/s with a thickness of 50 nm. In the asymmetric configuration, Au electrode was deposited by thermal evaporation using a proper shadow mask, and low-WF LiF/Al electrode was successively deposited using a proper shadow mask after a brief vacuum break: for the Au electrode a deposition rate of 2–4 Å/s with a thickness of 50 nm, and for the LiF/Al electrodes deposition rates of 0.1–0.2 and 1–2 Å/s with thicknesses of 0.7 and 70 nm, respectively. Both OLEFETs had a long channel width (*W*) of 10 mm with a channel length (*L*) of 200 μm.

Characterization of the morphological and structural properties of the active layers grown on the SiO₂ substrates was performed using AFM (PSI Co.) and XRD (Rigaku Co.). Unlike most of the former experiments carried out in a vacuum and/or under an inert atmosphere, all the *I*-*V* and *I*-*V*-*L* characteristics of the devices in this study were simultaneously measured *in air* using an optical probe connected to an HP4145B, and a Newport 818-UV Si photodiode with an 1830-C power meter. LSCM images and PL spectra of the single- and bi-layers were obtained using a Carl Zeiss LSM700 laser scanning confocal microscope and Hitachi F7000, respectively. The light emission images of the OLEFETs were obtained using a CCD (Q Imaging Co.) camera mounted on an optical microscope at the probe station.

3. Results and discussion

3.1. Surface morphological and structural properties

Comparative analysis of the surface morphological and structural properties for organic active layers deposited on the SiO₂ substrates was performed using AFM, XRD, optical spectroscopy

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