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Hybrid flexible ambipolar thin-film transistors based on pentacene and ZnO capable of low-voltage operation

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A B S T R A C T

A flexible ambipolar thin-film transistor (TFT) was developed that consisted of a pentacene layer and a ZnO layer modified with a dodecanoic acid self-assembled monolayer on a flexible polyarylate substrate. The TFT exhibited balanced hole and electron mobilities of 0.3 and 0.2 cm^2 V^{-1} s^{-1} and showed flexible thin-film characteristics. Additionally, because the anodized Al_2O_3 dielectric layer is a high κ material compared with a normal SiO_2 dielectric layer, the flexible ambipolar TFT was able to operate at a low voltage of 5 V.

Thin-film transistors (TFTs) based on organic semiconductors or metal oxide semiconductors have attracted considerable attention due to their potential utility in low-cost, easily processable, and flexible electronic devices that could be used as potential alternatives to transistors based on conventional silicon technologies [1]. To date, many types of organic semiconductors for TFTs have been synthesized and described [2]. Organic semiconductors are generally used in unipolar devices, and exhibit mostly *p*-type semiconducting behavior [3].

Although various *p*-type semiconductors with impressive mobilities have been reported, the development of *n*-type organic semiconductors suitable for use in TFT components of complementary logic circuits has been limited because of the difficult chemistry involved in the synthesis of materials with high stability in the presence of atmospheric water and oxygen [4]. Among metal oxide semiconductors, such as zinc oxide (ZnO), indium zinc oxide, zinc tin oxide and indium gallium zinc oxide, most show *n*-type semiconducting behavior and have the advantages of high electron mobility, optical transparency, and excellent environmental stability [5,6].

Ambipolar TFTs have also been intensively studied both for fundamental research and for technological applications in complementary logic circuits, which can be used to overcome the limitations of integrated circuits that use only unipolar logic [7,8]. In addition, complementary circuits based on ambipolar TFTs can be fabricated using less complex processes and at lower cost compare to unipolar TFTs [9]. Ambipolar TFTs based on organic semiconductors with various types of layer structures, such as bilayer, blend, and single-component structures, have been reported by many groups [8,10].

Our group recently developed high-performance ambipolar TFTs and an inverter based on pentacene/self-assembled monolayer (SAM) modified ZnO hybrid structures with well-balanced hole and electron mobilities [4]. In that previous

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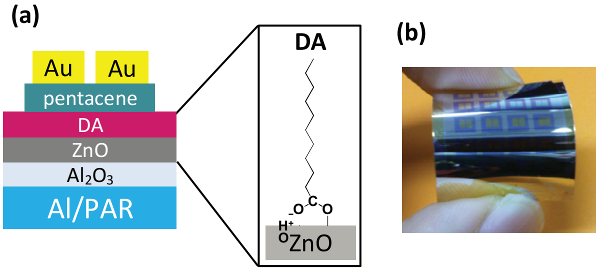

Fig. 1. (a) Schematic diagram of the ambipolar TFT based on a pentacene/ZnO hybrid structure in which a DA SAM was inserted at the interface between the pentacene and ZnO, and the chemical structure of a DA molecule bound to the ZnO active layer; and (b) a photograph of the flexible TFT devices on a PAR substrate.

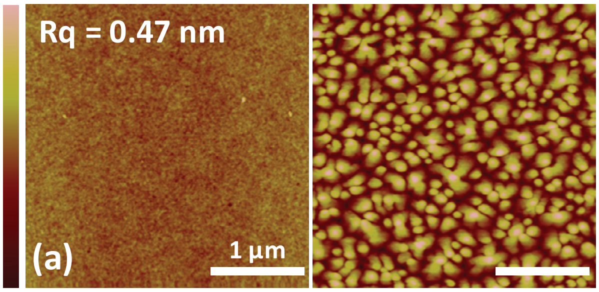

Fig. 2. Height-mode AFM images of (a) the bare ZnO and (b) the pentacene film. The vertical scales in (a) and (b) represent 10 and 50 nm, respectively.

report we fabricated the TFT devices on rigid silicon substrates; hence, the suitability of the TFTs for use in flexible electronic devices could not be tested. For TFTs to be suitable for use in flexible electronic devices, they must be able to withstand frequent flexing [11]. Here, we present ambipolar TFTs based on pentacene and ZnO with a flexible polyarylate (PAR) film as the substrate for use in flexible electronic devices.

Fig. 1 shows a schematic diagram of an ambipolar TFT based on an organic–inorganic hybrid structure composed of pentacene as the upper *p*-type layer and ZnO as the lower *n*-type layer. The PAR substrate was a 200-µm-thick film of AryLite^{TM} A200HC (Ferrania Technologies). Aluminum (Al) gates of thickness 200 nm were deposited onto the PAR film by thermal evaporation. Following Al deposition, anodization was performed at a constant current density of 0.32 mAcm^{-2} and a voltage of 30 V in a 0.05 M ammonium pentaborate octahydrate electrolyte solution at 25 °C in order to form a 100-nm-thick Al_2O_3 insulator layer. The ZnO thin film used as an *n*-type channel material was deposited from $Zn(OH)_2$ in aqueous ammonia solution (0.1 M, pH 13.5) by spin coating in order to form a 20 nm thick ZnO thin film. Before spin coating, the prepared solution was vigorously stirred for 12 h at room temperature and filtered through a 0.2 µm membrane filter. The coated ZnO layer was annealed at 300 °C for 30 min. The PAR substrate had a high thermal stability and was not affected by this annealing step [12]. Because of the poor compatibility between pentacene and ZnO, a dodecanoic acid (DA) SAM was deposited on the ZnO active layer prior to deposition of the pentacene layer. To achieve this, the ZnO layer was first ultraviolet–ozone-treated for 15 min and then a 1 mM solution of DA in ethanol was spin-coated onto the ZnO, resulting in the formation of a monolayer of DA through chemical bonding between the carboxylic acid groups of DA and ZnO. The DA-treated ZnO was then spin-coated again using pure ethanol to remove physically adsorbed DA molecules. After drying, a 50-nm-thick pentacene film was deposited onto the SAM-treated ZnO layer at room temperature, under a vacuum pressure of $\sim 10^{-6}$ Torr with a deposition rate of 0.2 Ås^{-1}. Finally, 80-nm-thick Au source and drain electrodes were deposited though a shadow mask. The channel length and width were 100 and 1500 µm, respectively. The electrical characteristics of the TFTs were measured in air at room temperature using Keithley 2400 and 236 source/measurement units.

For applications in electronic devices, ambipolar TFTs must have balanced hole and electron mobilities. To fabricate a high-performance ambipolar TFT based on a pentacene/ZnO bilayer structure with balanced hole and electron mobilities and a high hole mobility (μ_h), the pentacene layer must have high crystallinity; however, pentacene crystals grown on inorganic oxide films with rough surfaces and high surface energies typically exhibit low crystallinity [5,13]. In the present work, atomic force microscopy (AFM) in tapping mode was carried out to examine the morphology of the bare ZnO film. As shown in Fig. 2, the ZnO surface was smooth (root-mean square roughness (R_q) ∼ 0.47 nm). In previous report, the pentacene film on ZnO surface without SAM layer yielded the low hole mobility (6.3×10^{-3} cm^2 V^{-1}s^{-1}) because the compatibility between the pentacene and the ZnO is not good, which causes inferior growth of pentacene crystals [14]. To further improve the surface for pentacene deposition, we introduced a DA SAM at the interface between the pentacene and ZnO layers based on previous findings showing that the inclusion of such a SAM enabled the control of the surface energy and the permanent dipole field at the interface and produced well balanced ambipolar behavior [4]. In addition, previous work in our group has