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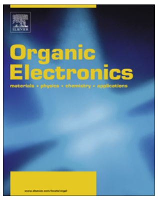

Environmental effects on the electrical behavior of pentacene thin-film transistors with a poly(methyl methacrylate) gate insulator

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ABSTRACT

The electrical properties of top-contact pentacene thin-film transistors (TFTs) with a poly(methyl methacrylate) (PMMA) gate dielectric were analyzed in air and vacuum environments. Compared to the vacuum case, the pentacene TFT in air exhibited lower drain currents and more pronounced shifts in the threshold voltage upon reversal of the gate voltage sweep direction, together with a decrease in the field-effect mobility. These characteristic variations were explained in terms of two distinctive actions of polar H_2O molecules in pentacene TFT. H_2O molecules were suggested to diffuse under the source and drain contacts and interrupt the charge injection into the pentacene film, whereas those that permeate at the pentacene/PMMA interface retard hole depletion in and around the TFT channel. The diffusion process was much slower than the permeation process. The degraded TFT characteristics in air could be recovered mostly by storing the device under vacuum, which suggests that the air instability of TFTs is due mainly to the physical adsorption of H_2O molecules within the pentacene film.

1. Introduction

Organic thin-film transistors (TFTs) have attracted considerable attention from both academia and industry because of their potential for low-cost manufacture and compatibility with glass or plastic substrates. In recent years, the performance of organic TFTs (e.g. field-effect mobility, threshold voltage and subthreshold swing) has improved significantly. Some devices currently have superior characteristics to those of amorphous silicon transistors [1–3]. Prototype applications based on organic transistors include active-matrix displays, radio-frequency identification tags and various sensors [4–9]. On the other hand, the reliability of organic TFTs is a key factor for full-scale applications in commercial electronics.

With regard to the electrical stability, a continuous bias stress on the organic TFT causes a large shift in the threshold voltage and hysteresis in the transfer characteristics. As a consequence, the electrical properties of the organic TFT deteriorate. Because such bias-stress-induced instabilities are caused mostly by charge injection into the gate dielectric and/or fixed charges formed at the organic semiconductor/gate dielectric interface [10–12], chemical modifications to the dielectric materials and dielectric surfaces are essential for enhancing the electrical stability of organic TFTs [13,14]. Organic transistors also degrade when exposed to ambient air, sometimes exacerbating the bias-stress instability [15–20]. This should be distinguished from the effects of exposure to volatile chemicals on both

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the TFT characteristics and the bias-stress effect [21]. Previous studies on this topic highlighted the detrimental effects of atmospheric moisture (H_2O) and oxygen (O_2) on organic TFTs. Polar H_2O molecules capture the charges in the conducting channel of the transistor, which decrease the saturation current and field-effect mobility [22,23]. Oxygen molecules can oxidize organic semiconductors that have narrow band gaps and high levels of the highest occupied molecular orbital, which impedes charge transport in organic TFTs [24–26].

The surface morphology and crystallinity of organic semiconductor films are correlated intimately with the environmental stability of the associated transistors. For example, Weitz et al. reported that the grain boundaries in the polycrystalline organic layer are responsible for the mobility degradation in the transistor under ambient air conditions [27]. Simeone et al. suggested that a greater density of pentacene grains leads to higher sensitivity to H_2O and an increase in the hysteresis of the transfer characteristics of pentacene TFTs [28]. Bae et al. reported also that cracks that develop in the solution-processed organic film act as trapping sites for H_2O and/or O_2, thereby degrading the TFT performance [29]. This suggests that the denser packing of organic molecules and fewer grain boundaries on the surface can help enhance the air-stability of organic TFTs by providing a kinetic barrier to the diffusion of ambient oxidants and reducing the invasion pathway of air H_2O and O_2 molecules [30–32]. Furthermore, a range of encapsulation approaches have been suggested to protect these transistors from air exposure [33–35]. In certain cases, the molecular structure of organic semiconductors is modified chemically through the judicious choice of conjugated units and side chains to improve their air stability [30,32,36–38].

Nevertheless, the realization of a fully air-stable organic transistor is still a key challenge. The problem stems from an insufficient understanding of the behavior of air molecules in organic TFTs. Although considerable effort has focused on explaining environmentally-influenced variations in the organic TFT properties in terms of the traps induced at the organic semiconductor/gate dielectric interface and energetic states generated within the semiconductor band gap, the effects of air molecules on the charge movement in organic TFTs has received less attention. This study focused on the environmental effects of the charge injection and transport characteristics of top-contact pentacene TFTs with a poly(methyl methacrylate) (PMMA) gate dielectric. The space-charge-limited transport in the pentacene film and the capacitance–voltage characteristics of the pentacene transistor were analyzed in air and vacuum environments. The threshold voltage and field-effect mobility were also examined upon reversal of the gate voltage sweep direction.

2. Experimental

The pentacene TFTs were fabricated on a glass substrate; Fig. 1 shows the device architecture. A stripe-patterned 40-nm-thick Al gate electrode was evaporated thermally onto a substrate through a shadow mask. For the gate insulator, a PMMA film was formed by spin-coating from solution (Sigma–Aldrich, approximately 4.8 wt.% dissolved in anisole) and curing for 60 min at 120 °C; the resulting thickness was approximately 150 nm. Pentacene (Sigma–Aldrich and used as received) was then evaporated thermally onto the insulator-coated substrate to a thickness of 30 nm through a second shadow mask. The substrate was kept at room temperature and the deposition rate was approximately 0.05 nm/s. Finally, the top-contact pentacene TFT was completed by depositing 40-nm-thick Au source (S) and drain (D) electrodes through a third shadow mask. The channel length and width were 200 μm and 4000 μm, respectively. The device was stored in the dark and air for 12 h. Subsequently, electrical characterization of the transistor was carried out in the dark and in either air or a vacuum using an HP 4140B picoammeter/voltage source. The capacitance versus voltage behavior was monitored using an HP 4192A impedance analyzer.

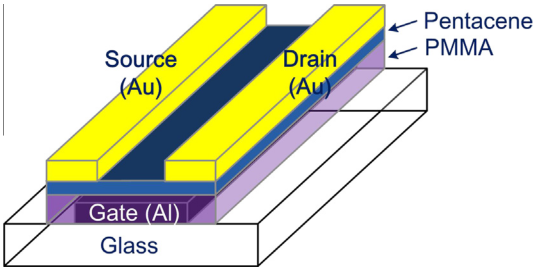

Fig. 1. Schematic diagram of a top-contact pentacene TFT.

3. Results and discussion

3.1. Output characteristics and total resistances of the pentacene TFT

The output characteristics of the pentacene TFTs were measured by varying the drain voltage (V_D) from 0 to −50 V in −0.5 V increments, at a constant gate voltage (V_G) of −10 V. The output curve was first obtained in air. A second set of data was recorded after keeping the device under a vacuum of approximately 10^{-2} mbar for 30 min. A third experiment was then performed within 10 s after breaking the vacuum, followed by a fourth set of measurements after 3 h in air. The relative humidity of ambient air was in the range of 40–50%. The resulting output characteristics of the pentacene transistor show typical *p*-channel operation with excellent saturation behavior (Fig. 2a–c). The value of the saturated drain current obtained in air ($I_{D,sat}$, approximately −0.10 μA, Fig. 2a) increased by approximately 20% ($I_{D,sat}$, approximately −0.12 μA, Fig. 2b) after storing the device under vacuum. Previously, Wang et al. reported that pentacene transistors in air exhibit charge-trapping behavior, which reduces the drift velocity of holes, and a high vacuum environment helps enhance the device performance [39]. In agreement with this finding, the increase in $I_{D,sat}$ under a vacuum in these results also provides evidence of the detrimental effect of air molecules on the channel conductivity of pentacene TFT. On the other