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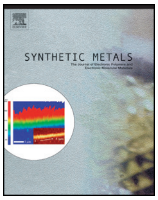

High performance low-voltage organic field-effect transistors enabled by solution processed alumina and polymer bilayer dielectrics

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ABSTRACT

High performance low-voltage pentacene-based organic field-effect transistors (OFETs) are fabricated utilizing solution-processed alumina (Al_2O_3) as high-k gate dielectric, and the effects of gate dielectric capacitance on the device performance were investigated. The results show that Al_2O_3 gate dielectric with optimized thickness can afford OFETs performance with field-effect mobility of 0.65 cm^2/Vs, threshold voltage of -0.6 V, I_{on}/I_{off} ratio of 4×10^3 and sub-threshold swing of 0.45 V/dec, at operating voltage as low as -4 V. After employing a very thin PMMA film onto Al_2O_3, the mobility of these OFETs with bilayer dielectric can be further boost up to 0.84 cm^2/Vs. According to electrical and microscope characterizations, the performance improvement of OFETs can be contributed to low surface trap density and high capacitance density of Al_2O_3/PMMA bilayer dielectrics.

1. Introduction

In the past decade, organic field-effect transistors (OFETs) have attracted considerable attention owing to their potential applications on micro-electronics, such as electronic papers, sensors, and large-area flexible displays [1–4]. To date, a great number of high performance OFETs have been developed, and most of their carrier motilities are comparable to amorphous silicon a-Si:H transistors [5,6]. However, one of the key problems of these high mobility OFETs is that high operating voltages are required for obtaining adequate channel currents. This is because the dielectrics most used in OFETs are organic semiconductor-compatible low-k materials, i.e., poly(methylmethacrylate) (PMMA), polystyrene (PS), and/or OTS-treated SiO_2, thus, additional energy losses may occurred if implanted those transistors into circuits [7–9].

Reducing film thickness of dielectric layers is an ordinary route to lower operating voltage. Nevertheless, thin polymer dielectric films will also cause gate leakage current increase [10]. High-k inorganic metal-oxides, such as TiO_2 [11], HfO_2 [12], ZrO_2 [13] and Al_2O_3 [14,15], are good candidates to substitute organic dielectrics for low voltage operation transistors. The corresponding examples are widely reported, especially in inorganic transistors [16,17]. Among these high-k metal oxide dielectrics, Al_2O_3 has attracted great attention owing to its good dielectric strength, large bandgap (E_g = 8.45–9.9 eV) and considerable dielectric constant (k = 7.0–9.0) [18,19]. Furthermore, Al_2O_3 exhibits many other advantages, such as low-cost, chemical stability, and solution processable [20–26]. Using bare Al_2O_3 as gate dielectrics in OFETs can somewhat lower operating voltage, but may not improve carrier mobility due to surface properties mismatch between organic semiconductors and Al_2O_3. As well known, structural ordering at organic semiconductor/dielectric interfaces dominates carrier transport in channels [27]. In contrast, low-k polymer dielectrics can provide smooth, low trap density surface, enabling effective carrier transportation at such interfaces [27–30]. Therefore, using metal oxide/polymer bilayer dielectrics is a promising route to conquer these problems [31–33]. However, the conventional deposition methods of metal oxide for OFETs are expensive, such like chemical vapor deposition (CVD), atomic layer deposition (ALD) and radio-frequency (RF) magnetron sputtering [34–36]. To realize low cost roll-to-roll fabrication, solution processed is highly desired.

In this work, we fabricated the pentacene-based OFETs with solution processed Al_2O_3 as gate dielectric, and for first time investigated the effects of the gate dielectric capacitance of solution processed Al_2O_3 on OFETs device performance. The OFETs show a field-effect mobility of 0.65 cm^2/Vs, a threshold voltage of -0.7 V, a drain current on/off ratio of 4×10^3 and a sub-threshold swing of 0.45 V/dec. On the other hand, we combined solution

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processed Al_2O_3 and low-k polymer PMMA to provide compatible surface for organic layer growth [29]. The corresponding OFETs with bilayer dielectrics exhibit performance improvement in key properties such as field-effect mobility, threshold voltage, and sub-threshold swing. The results imply that increase capacitance of dielectric and modify insulating layer can be used in improving the performance of OFETs.

2. Experimental

Aluminum oxide (Al_2O_3) sol was prepared by dissolving aluminum nitrate nonahydrate ($Al(NO_3)_3 \cdot 9H_2O$, 99.99%, Sigma–Aldrich) into 2-methoxylethanol in a concentration ranging from 0.1 to 0.5 M and then stirred for 12 h in ambient conditions. PMMA solution was prepared by dissolving PMMA (Sigma–Aldrich, average MW ~120,000) into anisole with a concentration of 0.5 wt%, and then stirred for 12 h in ambient conditions.

Fig. 1 shows the device fabrication process, before dielectric layer deposition, indium tin oxide-coated glass substrates were cleaned successively in an ultrasonic bath with detergent, acetone, deionized water and isopropanol for 15 min each. The Al_2O_3 layer was deposited by spin-coating the Al_2O_3 sol onto the cleaned indium tin oxide-coated glass substrates at 3000 r/min for 40 s, and annealed on a hot plate at 200 °C for 30 min. Subsequently, the modify layer was deposited by spin-coating the 0.5 wt% PMMA precursor solution onto the Al_2O_3-coated substrates and then baked at 80 °C for 30 min. Next, 50 nm pentacene (Sigma–Aldrich without purification) was evaporated onto the dielectrics in high vacuum chamber (pressure $\sim 2 \times 10^{-4}$ Pa) at a deposition rate of 0.2–0.3 Å/s. Finally, 50 nm Au electrodes were thermally evaporated using a metal shadow mask at a rate of 0.2 Å/s under 3×10^{-3} Pa. The width and length of the channel on the shadow mask were 100 μm and 1 cm. As for leakage and capacitance characterization, a metal–insulator–metal (MIM) structure was fabricated by direct deposition of 30 nm thick Ag onto the Al_2O_3 and Al_2O_3/PMMA dielectric.

The electrical characteristics of OFETs were measured using a Keithley 4200-SCS in ambient conditions. The mobility (μ) was calculated in the saturation region of transfer curves using Eq. (1):

$$I_{DS} = \left(\frac{W}{2L}\right)\mu C_i (V_{gs} - V_{th})^2 \qquad (1)$$

where C_i is the capacitance per unit area of the insulator, V_{th} is the threshold voltage, and V_{gs} is the gate voltage. W and L are the channel width and length.

The morphologies of Al_2O_3 dielectrics and Al_2O_3/PMMA layer were characterized by atom force microscope (AFM) in tapping mode, the thicknesses of the insulators were measured via X-ray reflectivity (XRR), the frequency-capacitance measurement of the MIM structure was performed by 4294A (Santa Clara, CA, USA) in a frequency range of 100 Hz–1 M Hz.

3. Results and discussion

Fig. 2(a–e) shows the tapping mode AFM images of the Al_2O_3 films with various thickness, the Al_2O_3 insulators exhibit a homogenous and smooth surface, with root-mean-square (RMS) roughness of 0.35, 0.30, 0.29,0.27 and 0.26 nm for Al_2O_3 film thickness of 35, 56, 73, 87 and 102 nm. For films with roughness <0.5 nm, the dielectric roughness becomes an assisting factor in determining mobility [37]. In addition, as shown in Fig. 2f, the RMS roughness of PMMA/Al_2O_3 films is 0.26 nm, which implied that modification treatment would not affect the surface roughness of Al_2O_3 dielectric. The smooth surface of the Al_2O_3 films and PMMA modified dielectrics exhibits the potential for high-performance OFETs. The X-ray diffraction (XRD) characterization is conducted on the solution-processed single-layer Al_2O_3 system. As can be seen in Fig. 2g, the absence of diffraction peaks in the XRD patterns indicates that the amorphous nature of the oxide dielectrics obtained at low processing temperature. The amorphous feature of dielectrics is favorable for transistor application, since it has been proven that amorphous oxide dielectrics enable low-leakage current and decrease surface roughness in some case [38].

The capacitance of Al_2O_3 films with different thicknesses are shown in Fig. 3a. It can be observed that the capacitance of Al_2O_3 decrease with increasing thickness, which is consistent with the theoretical of parallel-plate capacitors. Meanwhile, the bare Al_2O_3 dielectrics breakdown when frequency goes higher than 10^5 Hz, indicated the existence of high leakage current. The dielectric constant of Al_2O_3 at 10 kHz was determined to be 7.2 ± 0.2 (calculated from the Eq. (2):

$$C_{ox} = \frac{\varepsilon_0 k}{d} \qquad (2)$$

where ε_0 is the permittivity in vacuum, d is the thickness, and k is the dielectric constant). Shown in Fig. 3b, after the modification of PMMA, the total capacitance decreased. The double layer of Al_2O_3 (56 nm)/PMMA (10 nm) exhibited quite a high capacitance value

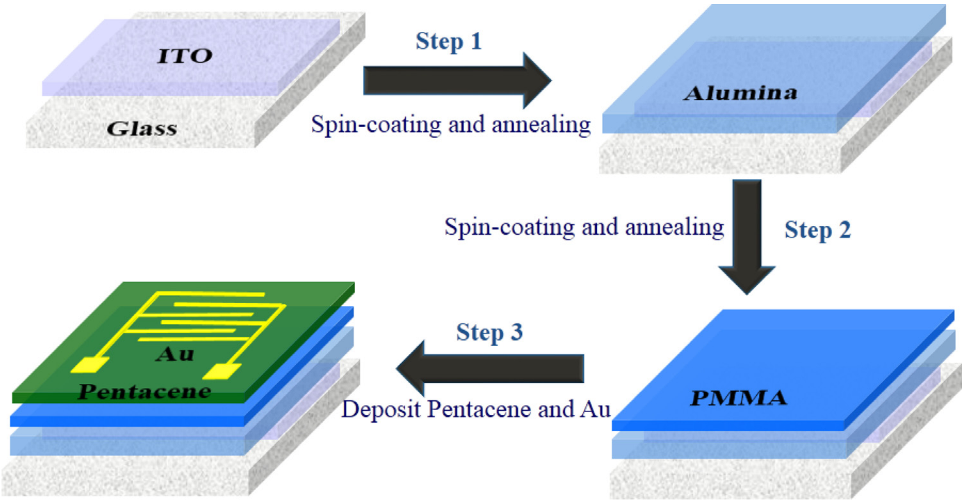

Fig. 1. Schematic of the fabrication process.