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Letter

Low-voltage organic thin film transistors with hydrophobic aluminum nitride film as gate insulator

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Abstract

This paper reports on the low-voltage (<5 V) pentacene-based organic thin film transistors (OTFTs) with a hydrophobic aluminum nitride (AlN) gate-dielectric. In this work, a thin (about 50 nm), smooth (roughness about 0.18 nm) and low-leakage AlN gate dielectric is obtained and characterized. The AlN film is hydrophobic and the surface free energy is similar to the organic or the polymer films. The demonstrated AlN–OTFTs were operated at a low-voltage (3–5 V). A low-threshold voltage (−2 V) and an extremely low-subthreshold swing (~170 mV/dec) were also obtained. Under low-voltage operating conditions, the on/off current ratio exceeded 10^6, and the field effect mobility was mobility was 1.67 cm^2/V s.

Keywords: OTFTs; AlN; Pentacene; High-*k*; Sputtering; Contact angle; Surface energy

1. Introduction

Organic thin-film transistors have been proposed for use in various applications in displays and flexible electronics [1,2]. However, the high operating-voltage remains a limitation on organic transistors. Capacitance is important in lowering the operating voltage. A larger capacitance is well known to accumulate more carriers and turn on transistors at lower voltage. Two approaches have been proposed to increase the gate capacitance: to reduce the thickness of the gate-dielectric [3] and to use a high-*k* material as a gate-dielectric [4]. Nevertheless, controlling gate leakage is an additional difficulty [5]. As well as the increase in capacitance, the surface polarity (hydrophilic or hydrophobic) of the gate-dielectric is an important factor. Many researchers have shown that polymer dielectrics are suitable for organic film deposition because they have similar surface energies to those of organic films. The

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dielectric polarity is altered by the self-assembled monolayer (SAM) on inorganic dielectrics. Inorganic dielectrics with lower surface energy offer improved device performance; such surfaces reduce the number of interface traps in OTFTs [6,7].

This work presents AlN film as the gate dielectric for OTFTs. The AlN film is known to have high chemical and physical stability, as well as high dielectric permittivity [8]. In this study, the AlN film was hydrophobic and the surface energy was similar to that of the pentacene film. Moreover, the AlN film can be deposited at 150 °C by using RF-sputtering system. High breakdown voltage and low-leakage-current can be obtained. This letter also presents the favorable switching characteristics of AlN–OTFTs.

2. Experimental

AlN film was deposited at low-temperature (150 °C) by a radio-frequency (RF) sputtering system [9]. For AlN-deposition, the n^+-Si wafer was used as the substrate. The wafer was rinsed in the deionization water, and was then immersed in the acetone with ultrasonic clean. Consequently, the wafer was dipped in dilute HF solution ($HF:H_2O = 1:100$) to remove the native oxide from the Si wafer. Finally, the wafer was transferred to the RF-sputtering system immediately. The system was then pumped down to a base pressure of under 2×10^{-6} Torr before gas was admitted. Mixed argon and nitrogen gas was monitored by mass flow controllers (MFC). The AlN films were deposited at a total pressure of 2.5 mTorr. All the relevant experimental details have been published elsewhere [9]. After the AlN films were deposited, pentacene film was deposited through the shadow mask. The pentacene material obtained from Aldrich without any purification was directly placed in the thermal coater for deposition. The substrate was heated to 70 °C during the deposition at a pressure of around 1×10^{-6} Torr. The thickness of the pentacene film was about 100 nm and the deposition rate was around 0.5 Å/s, monitored by a quartz crystal oscillator. Then, Au was deposited as the source/drain electrodes on the pentacene film. The thickness of the electrode pad was 1000 Å. The channel width and length were defined as 600 μm and 100 μm. Metal–insulator–semiconductor (MIS) – Au/AlN/Si was also fabricated to analyze the gate leakage and the dielectric properties. The area of the Au pad was $500 \times 500\ \mu m^2$. All electrical characteristics were measured using Agilent 4156 and Agilent 4284 analyzers.

3. Results and discussion

In our previous investigation, the AlN film was deposited at a higher substrate temperature [9]. The AlN film has a grain-structure and is highly *c*-axis oriented. If the AlN film is deposited at high temperature to form the gate-dielectrics, the grain boundaries will affect the surface roughness and reduce the uniformity. The grain boundaries commonly serve as leakage paths. Since the dielectric roughness and leakage are critical in OTFT fabrication, in this work, AlN was deposited at a lower substrate temperature (150 °C). Fig. 1(a) reveals the AFM image of the 150 °C AlN film. Unlike a high-temperature AlN film [9], the 150 °C AlN film was smooth. The surface roughness was only 0.18 nm. Fig. 1(b) presents the scanning electron microscopic (SEM) image to verify the dielectric quality. In the 150 °C sputtered AlN film (with a thickness of around 50 nm), no significant surface irregular and pinholes were observed. The fluctuation of dielectric thickness is appeared; it may result in performance variation when OTFT size is reduced to micro-scale. The 150 °C AlN film had favorable surface properties. The dielectric properties were also examined. The measured capacitance of the MIS structure was approximately 104 nF/cm^2. Fig. 1(c) shows the gate-leakage, which was as low as $\sim 10^{-8}$ A/cm^2 in an electric field of 1 MV/cm.

In this investigation, OTFTs were fabricated with thin AlN film (thickness is about 50 nm, dielectric constant is about 6) as gate-dielectric. Fig. 2(a) presents the transfer characteristics. The AlN–OTFTs can be operated at a relatively low-voltage (~5 V). The on/off current ratio was about 10^6; the threshold voltage was only −2.1 V; the field effect mobility was 1.67 cm^2/V s. The subthreshold swing was 170 mV/decade. The magnitude approached the theoretical minimum, ~60 mV/decade $(kT/q \cdot \ln(10))$ [7]. Fig. 2(b) shows the output characteristics, the AlN–OTFT were operated in the saturation region at low-drain bias (~3 V). Since the subthreshold swing represents the interface quality and the trap behavior, [10] the maximum interface trap density is given by the approximation, [11]

$$N_{SS} = \left[\frac{S \cdot \log(e)}{kT/q} - 1\right] \cdot \frac{C_i}{q}$$

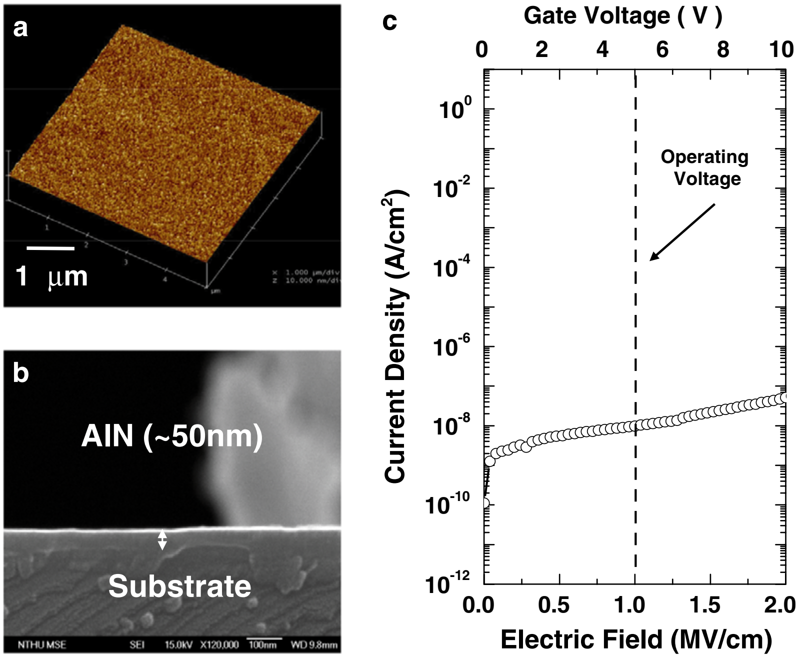

Fig. 1. (a) The AFM image shows the 150 °C AlN film, and the scanning size is 5 × 5 μm^2. (b) The SEM image is the cross-section view of the AlN film on substrate. (c) The leakage current of the Au–AlN–Si structure as a function of the electric field and gate voltage.

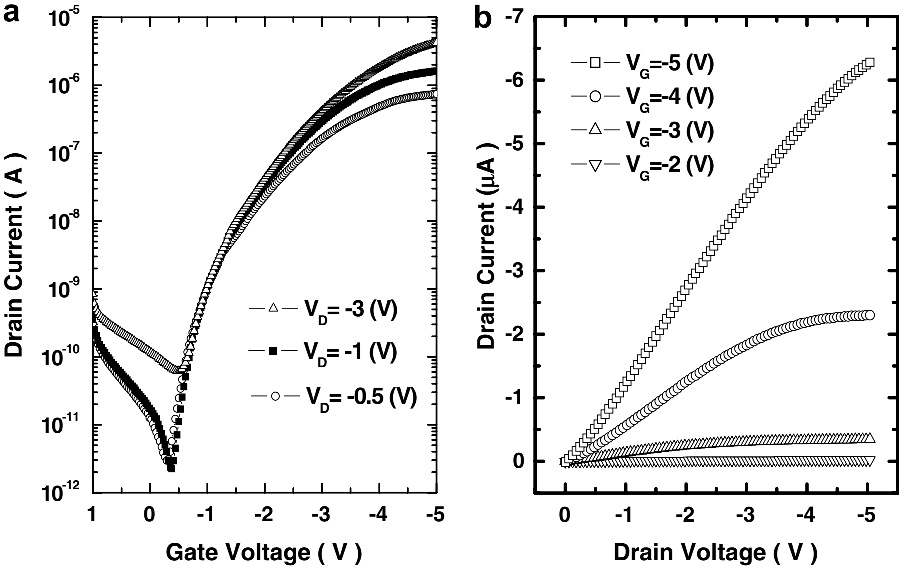

Fig. 2. (a) The transfer characteristics of OTFTs with AlN gate dielectric. (b) The corresponding output characteristic.

where S is the subtheshold swing; C_i is the capacitance per unit area; k is Boltzmann's constant, and T is the absolute temperature. Substituting $C_i = 104$ nf/cm^2 and $S = 170$ mV/decade yields an approximate interface trap density in the devices of 1.2×10^{12} cm^{-2} eV^{-1}. To the authors' limited knowledge, this value is close to the lowest reported for organic transistors [3,7,12].

The organic/dielectric interface defects strongly affect the performance of OTFTs. Surface polarity is key to reducing the interface defect. SAM-treatment and a polymer dielectric are widely employed to change the polarity of the surface. Researchers have recently demonstrated that interface defects can be minimized by changing the dielectric polarity [6,7,13]. Accordingly, the AlN surface polarity was determined herein. The modified Fowkes' equation for the surface polarity in terms of the surface energy is introduced [14]

$$(1+\cos\theta)\gamma_{\rm L} = 2(\gamma_{\rm S}^{\rm d}\gamma_{\rm L}^{\rm d})^{1/2} + 2(\gamma_{\rm S}^{\rm p}\gamma_{\rm L}^{\rm p})^{1/2}$$

where θ is the contact angle between probing liquid and the solid surface; $\gamma_{\rm L}$ is the total surface free energy of the probing liquid; $\gamma_{\rm L}^{\rm d}$ is the dispersion component, and $\gamma_{\rm L}^{\rm p}$ is the polarity component. Based on this approximation, the total surface free energy of the solid surface is,

$$\gamma_{\rm S} = \gamma_{\rm S}^{\rm d} + \gamma_{\rm S}^{\rm p}$$

where the total surface free energy $\gamma_{\rm S}$ is the sum of $\gamma_{\rm L}^{\rm d}$ (dispersion component) and $\gamma_{\rm L}^{\rm p}$ (polar component). The surface free energy can be calculated by measuring the contact-angle between the solid surface and the different probing liquids. As shown in Fig. 3(a)–(c), the optical images of various liquid drops on the 150 °C AlN films were captured using a CCD camera. The water–AlN, ethylene glycerol–AlN and di-iodo-methone–AlN contact angles were 72.9° ± 5°, 60.7° ± 4° and 43.8° ± 2°, respectively. The Owens-Wendt-Rabel and Kaelble's method [14] yields an estimated surface energy of AlN of 38.3 (mJ/m^2). As stated in Table 1; compared to the inorganic dielectrics, the AlN had a low-surface free energy [15,16]. The surface free energy was unusually lower than those of polymer dielectrics [17,18], pentacene, [19] and the HMDs-treated SiO$_2$ [20]. It was close to that of OTS-treated SiO$_2$ [20]. The particular characteristic differs from those of the other ceramic-based dielectrics. According to Chou et al. investigation [21], the deposited pentacene film is composed of both orthorhombic and triclinic pentacene. The "dielectric" surface free energy matched to the "orthorhombic pentacene film" (38 mJ/m^2) is the key factor to the high mobility OTFTs. It is believed that the voids and the

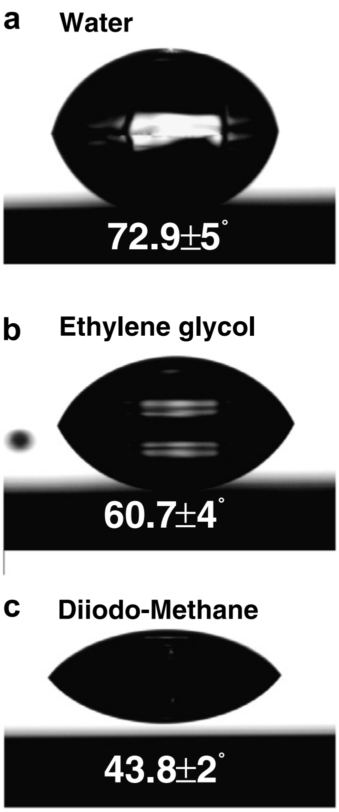

Fig. 3. (a) The water, (b) ethylene glycol, and (c) di-iodo-methane drops on the 150 °C AlN film for the surface-energy measurement. Each value below the drop indicates the contact-angle between the liquid drop and the AlN film. The entire optical images were captured from the CCD camera.

Table 1
Contact-angle measurements and the corresponding surface free energy of the commonly used dielectrics in the OTFTs fabrication

Substrate	Contact angle between liquid and sample			Surface free energy (mJ/m^2)	Reference
	D.I. water	Glycerol	Di-iodo-methane		
Al$_2$O$_3$	20–37			68–78	[15]
Si$_3$N$_4$	20–30			55–60	[16]
SiO$_2$	35.7	22.4	25.1	60	[20]
PVA	53.9	54		46.3	[17]
HMDs + SiO$_2$	53.7	53.7	43.9	45.4	[20]
PVP-copolymer	50–60			42	[18]
Pentacene				38,42–48	[19], [21]
AlN	72.9 ± 5	60.7 ± 4	43.8 ± 2	38.3	This work
OTS + SiO$_2$	78.9	81.8	43.9	34.9	[20]

incompletely stacked layers, which limited the carrier transport in the pentacene film, are reduced due to the match of surface energy [22]. In this study, the surface free energy of AlN agrees very well with that of the orthorhombic pentacene film, but not to the triclinic pentacene film [23]. Based on these arguments, the high performance AlN–OTFT is attributed to the match of orthorhombic pentacene film with AlN.

4. Conclusions

An AlN film was used as the gate dielectric for OTFTs. The pinhole free, smooth and extremely thin AlN film can be deposited by the RF-sputtering system at 150 °C. This low-temperature deposition process enables the potential application on polymer substrates. The dielectric leakage was significantly low, and the AlN has a surface free energy that is similar to that of the pentacene film. The transfer and output characteristics demonstrate that the AlN–OTFT has potential application in low-voltage and rapid-switching organic transistors.

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