Effects of Different Annealing Gases on Pentacene OTFT With HfLaO Gate Dielectric

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***Abstract*—Pentacene organic thin-film transistors (OTFTs) with HfLaO high-κ gate dielectric were fabricated. The dielectric was prepared by a sputtering method and then annealed in N_2, NH_3, O_2, or NO at 400 °C. The carrier mobility of the NH_3-annealed OTFT could reach $0.59\ \mathrm{cm^2/V \cdot s}$, which is higher than those of the other three devices. Moreover, the NH_3-annealed OTFT obtained the smallest subthreshold swing of 0.26 V/dec among them. Furthermore, $1/f$ noise measurement indicated that the NH_3-annealed OTFT achieved the smallest $1/f$ noise. All these should be attributed to the improved interface between the gate dielectric and the organic semiconductor associated with the passivation effects of the NH_3 annealing on the dielectric surface.**

***Index Terms*—Dielectric, high-κ, organic thin-film transistor (OTFT), $1/f$ noise.**

I. Introduction

ORGANIC thin-film transistor (TFT) (OTFT) has attracted wide attention in the academic and industrial fields [1], [2]. In comparison with its inorganic counterparts, OTFT has its own advantages. It can be fabricated on a flexible plastic substrate and can be realized in a large area at a low temperature. Moreover, since lithography is unnecessary, the cost can be greatly reduced. OTFT has potential to be applied to large-area flexible displays [3], RF-ID cards [4], electronic papers, and so on. Pentacene OTFT based on a SiO_2 dielectric has achieved a performance that is comparable to that of an amorphous-silicon TFT from the view of carrier mobility [5]. However, the operating voltage is usually too high (> 15 V) because of the low dielectric constant (κ) of SiO_2. The high operating voltage leads to large power consumption, which is particularly unacceptable for portable equipment. To solve this problem, an insulator with high κ value was prepared for the gate dielectric of OTFTs [6]–[8]. HfLaO has been reported to have less Fermi-energy pinning than HfO_2 in MOSFETs [9], [10], and the κ value of the former could be over 20. Furthermore, the subthreshold swing (SS) for OTFTs based on HfLaO was reported to be superior to that of polysilicon TFTs [11]. The carrier transport in the OTFTs is known to be decisively determined by the interface between the gate dielectric and the organic semiconductor. Therefore, in this letter, the effects of annealing the HfLaO gate dielectric in N_2, NH_3, O_2, or NO on pentacene OTFTs are studied, and it is found that the OTFT treated in NH_3 displays the best performance in terms of carrier mobility, SS, and $1/f$ noise characteristics.

II. Experimental Details

A silicon substrate (n-type, $\langle 100\rangle$, resistivity of $0.5-0.7\ \Omega \cdot$ cm) was cleaned according to the standard RCA method. Next, the wafer was dipped into 5% hydrofluoric acid to remove the native oxide. In the following, the wafer was washed by deionized water and dried in the common nitrogen stream. Subsequently, the wafer was inserted in the chamber of a sputterer (Denton Vacuum LLC Discovery 635). After the vacuum in the chamber was reduced to below 2×10^{-6} torr, a HfLaO film was prepared by RF sputtering (30 W) of a HfLa target ($\mathrm{Hf : La} = 6 : 4$ in molar ratio) in argon (24 sccm) and oxygen (3 sccm) to form the gate dielectric. Then, the samples were treated by annealing to improve the surface and bulk characteristics of the HfLaO film. The samples were annealed in N_2, NH_3, O_2, or NO, respectively, at a flow rate of 1000 mL/min and 400 °C for 10 min. Next, the back oxide of the wafer was removed with 20% hydrofluoric acid. Then, pentacene (purchased from Sigma-Aldrich) was evaporated on the dielectric by an evaporator (Edwards Auto 306). During the evaporation process, the substrate was kept at room temperature. The thickness of the pentacene film was 30 nm which was measured by a quartz-crystal oscillator, and the deposition rate was 1.2 nm/min. Finally, drain and source electrodes were formed by gold evaporation through a shadow mask at a vacuum 8×10^{-6} torr. The channel width and channel length of the shadow mask were 200 and 30 μm, respectively.

The I–V characteristics and noise characteristics of the OTFTs were measured by an HP 4145B semiconductor parameter analyzer, a Berkeley Technology Associates 9603 field-effect transistor (FET) noise analyzer, and an HP 35665A dynamic signal analyzer. The C–V characteristics of the Au–pentacene–HfLaO–Si structure were measured by an HP

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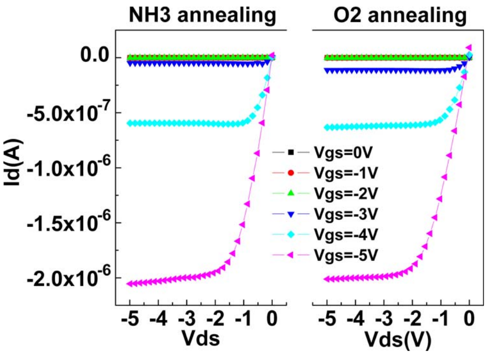

Fig. 1. Comparison of the output characteristics of the NH_3- and O_2-annealed OTFTs.

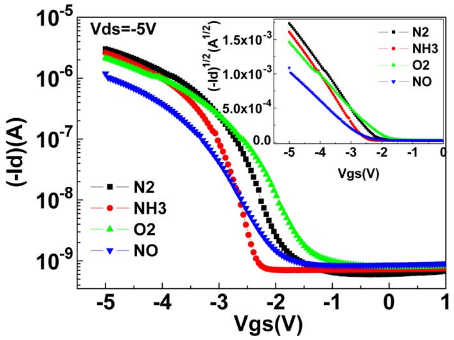

Fig. 2. Comparison of the transfer characteristics of the OTFTs. (Inset) $(-I_d)^{1/2}$–V_{gs} curve.

4284A precision LCR meter. All the electrical measurements were completed in a shielded probe station under ambient atmosphere. The thickness of the dielectric films was derived by a WVASE32 ellipsometer made by the J. A. Woollam Company, Inc.

III. RESULTS AND DISCUSSION

Fig. 1 shows the output characteristics of the OTFTs annealed in NH_3 or O_2. From these graphs, both OTFTs show good field-effect characteristics and could work well with a low supply voltage of 5 V.

Fig. 2 shows the comparison of the transfer characteristics of the OTFTs with dielectric treated in N_2, NH_3, O_2, or NO, respectively. The carrier mobility μ and threshold voltage V_{th} can be extracted from $(-I_d)^{1/2}$ versus V_{gs} in the saturation region for FETs [12]. The carrier mobility is given in (1), and the threshold voltage can be derived from the interception on the x-axis by extrapolating $(-I_d)^{1/2}$ versus V_{gs} to the x-axis

$$\mu = \frac{2L}{WC_{\mathrm{ox}}}\left(\frac{\partial\sqrt{-I_d}}{\partial V_{\mathrm{gs}}}\right)^2. \qquad (1)$$

The carrier mobility of the NH_3-annealed OTFT is 0.59 $\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$, which is higher than 0.49 $\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$ of the N_2-annealed OTFT, 0.34 $\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$ of the O_2-annealed OTFT, and 0.22 $\mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$ of the NO-annealed OTFT. The switching characteristics of the OTFTs can be evaluated by their SS. The NH_3-annealed OTFT obtains the smallest SS among the devices, thus the best switching characteristics.

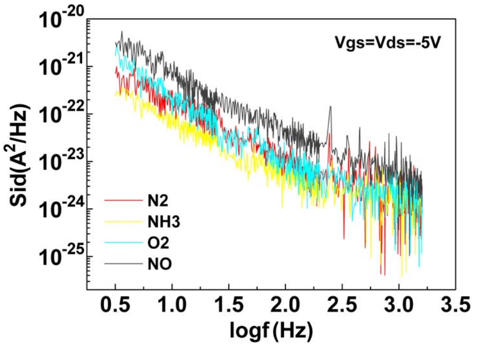

Fig. 3. Comparison of the $1/f$ noise spectrum of the OTFTs annealed in N_2, NH_3, O_2, or NO.

TABLE I
DEVICE PARAMETERS OF THE OTFTS AND CAPACITORS

	N_2	NH_3	O_2	NO
mobility (cm^2/Vs)	0.49	0.59	0.34	0.22
threshold voltage (V)	-2.27	-2.59	-2.01	-2.56
SS (V/dec)	0.34	0.26	0.45	0.60
on/off current ratio (10^3)	5.1	3.7	2.7	1.4
Id (μA) (Vds=Vgs=-5V)	3.01	2.60	2.14	1.19
Hooge's parameter α	0.035	0.021	0.086	0.440
tox (nm)	39.2	40.1	39.7	38.1
Cox (μF/cm^2)	0.228	0.230	0.218	0.228
k value	10.1	10.4	9.8	9.8

$1/f$ noise reveals the current fluctuation in the conduction channel and is closely related to the transport of carriers. Since organic devices may focus on low-frequency applications, $1/f$ noise is of significant concern due to the fact that noise power becomes larger in the lower frequency regime. Fig. 3 shows the comparison of the $1/f$ noise spectrum for the OTFTs. The NH_3-annealed OTFT has the least $1/f$ noise among the samples. According to the Hooge's expression for the $1/f$ noise of OTFTs [13], the Hooge's parameter α can be calculated as

$$\alpha = \frac{f^{\gamma} S_{\mathrm{id}}(f) L^2}{e\mu V_{\mathrm{ds}} I_d} \qquad (2)$$

where $S_{\mathrm{id}}(f)$ is the power spectral density and f is the measurement frequency. The NH_3-annealed OTFT has a Hooge parameter of 0.021, which is the smallest among the devices, hence the lowest noise level.

The device parameters of the OTFTs and capacitors are summarized in Table I. The higher carrier mobility, smaller SS, and lower $1/f$ noise level of the NH_3-annealed OTFT can be attributed to the improved interfacial characteristics between the organic film and the gate dielectric obtained by annealing the latter in NH_3. The reason should be that the hydrogen atoms decomposed from the NH_3 gas could passivate the dangling bonds at the surface of the HfLaO film and thus reduce the interface traps. This can be supported by Fig. 4, which shows the C–V characteristics (measured at 1-MHz frequency) of the Au–pentacene–HfLaO–Si unit with the dielectric annealed in N_2, NH_3, O_2, or NO. In Fig. 4, the NH_3-annealed unit displays the smallest hysteresis, while the NO-annealed structure has the largest hysteresis. The morphology of the pentacene film is shown in Fig. 5. From Figs. 4 and 5, the NO annealing is undesirable because it gives the largest hysteresis and smallest grains.

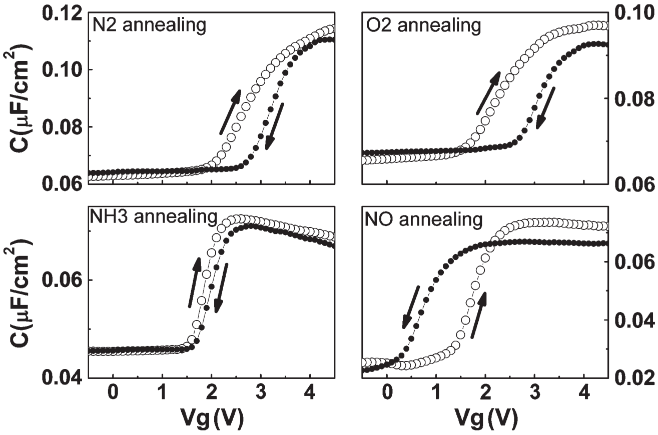

Fig. 4. C–V characteristics of the Au–pentacene–HfLaO–Si units.

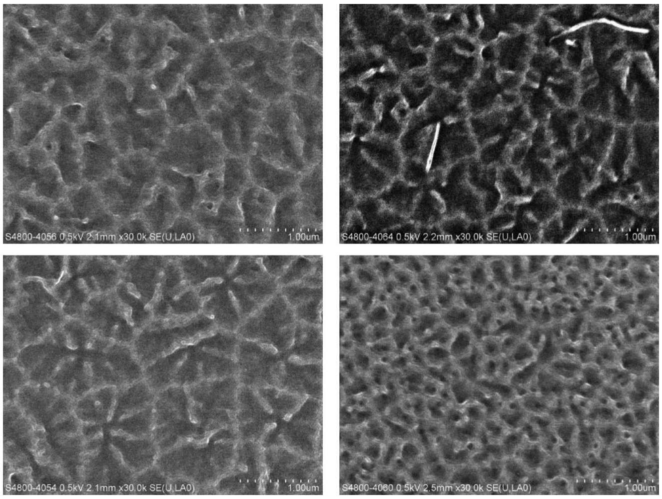

Fig. 5. SEM images of the pentacene film grown on the dielectric. (Northwest) N_2 annealing. (Northeast) O_2 annealing. (Southwest) NH_3 annealing. (Southeast) NO annealing.

In [11], pentacene OTFT with a more advanced structure was based on a HfLaO gate dielectric annealed in O_2, and its carrier mobility and SS were reported to be $0.71\ \mathrm{cm}^2/\mathrm{V}\cdot\mathrm{s}$ and 0.078 V/dec, respectively. Therefore, this study could provide a method to further improve the performance of the OTFT by annealing the HfLaO gate dielectric in NH_3. The OTFTs in this study display a lower on/off current ratio than that reported in [11]. One reason is that a patterned gate structure was used in [11]. Another possible reason is that the HfLaO film prepared by electron beam evaporation of compound HfLaO source [11] might achieve smaller dielectric leakage than our film made by sputtering of a metal target. Nevertheless, since our work is mainly related to the postdeposition treatment of the dielectric film, the device improvements mentioned previously should also be applicable to dielectric films fabricated by other techniques such as electron beam evaporation or atomic layer deposition.

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