Pentacene Thin-Film Transistors with HfO_2 Gate Dielectric Annealed in NH_3 or N_2O

L. F. Deng, W. M. Tang, C. H. Leung, P. T. Lai*, J. P. Xu, and C. M. Che

Abstract –**Pentacene-based Organic Thin-Film Transistor (OTFT) with HfO_2 as gate dielectric is studied in this work. The HfO_2 dielectric was prepared by RF sputtering at room temperature, and subsequently annealed in N_2O or NH_3 at 200 °C. The OTFTs were characterized by IV measurement and 1/f noise measurement. The OTFTs show small threshold voltage and can operate at as low as 3 V. Results indicate that the OTFT annealed in NH_3 shows higher carrier mobility, larger on/off current ratio, smaller sub-threshold swing and smaller Hooge parameter than the OTFT annealed in N_2O. Therefore, NH_3-annealed HfO_2 is a promising gate dielectric for the fabrication of high-performance OTFTs.**

I. INTRODUCTION

Organic thin-film transistors (OTFT) have got wide attention from researchers due to their advantages such as low cost and flexible substrate. They are considered as good candidates for applications like electronic paper, RFID tags, smart cards and large-sized flat-panel display [1], [2]. OTFTs with pentacene as active layer and SiO_2 as gate dielectric have achieved a performance which is comparable to amorphous Si thin-film transistors [3]. However, OTFTs with SiO_2 as gate dielectric often work at high operating voltage, which is commonly greater than 15 or 20 V. High operating voltage usually leads to high power consumption and causes inconvenience to the development of portable equipment. One way to decrease the operating voltage is to reduce the thickness of the gate dielectric [4]. However, too thin gate dielectric will cause large gate leakage. The other way to decrease the operating voltage is to replace the low-k SiO_2 with high-k dielectric [5], [6]. Hafnium (Hf) -based oxides (e.g. HfO2, HfON) are being actively investigated to act as the gate dielectric of inorganic transistors because of their better interface quality with the semiconductor [7], [8] as well as higher dielectric constant. The carrier transport in the OTFTs is decisively determined by the characteristics of the interface between the gate dielectric and organic semiconductor. In order to realize high-quality interface, one common method is to passivate the gate-dielectric surface by nitridation before the evaporation of organic layer. In this work, HfO_2 was prepared by sputtering method and then annealed in N_2O or NH_3 respectively to form the gate dielectric of pentacene OTFTs. The IV characteristics of the devices were measured, and used to deduce the field-effect carrier mobility, threshold voltage, sub-threshold swing and on/off current ratio. Moreover, 1/f noise was measured to evaluate the interface quality of the OTFTs.

II. EXPERIMENTAL DETAILS

Fig. 1 shows the cross-sectional view of the OTFT to be fabricated as follows. N-type <100> silicon wafers with a resistivity of 0.2 ~ 0.5 Ωcm were cleaned according to the standard RCA method. Then 5% hydrofluoric acid was used to remove the native oxide of the wafers. Subsequently the wafers were sputtered with a layer of HfO_2 as gate dielectric at room temperature. The sputterer was Denton Vacuum LLC Discovery 635. Before the sputtering, the vacuum in the chamber was kept below 2×10^{-6} Torr. The sputterer worked on RF (radio frequency) mode and the power was 30 W. The material of the target was HfO_2. Argon flowed in the reactive chamber at a rate of 24 sccm. In order to improve the interfacial characteristics, the samples were divided into two groups, each annealed in N_2O and NH_3 respectively with a gas flow rate of 1000 mL/min. The annealing process lasted for 10 minutes at a temperature of 200 °C. Hydrofluoric acid with 20% concentration was used to remove the back oxide of the silicon substrate which would be used as the gate electrode. Then pentacene (from Aldrich) was deposited on the dielectric in an Edwards Auto 306 evaporator. The substrate was not heated and the vacuum in the chamber was 4×10^{-6} Torr. The deposition rate was 1.1 nm/min and the final thickness of the pentacene film was 30 nm, which was detected by a quartz-crystal oscillator. Finally gold was evaporated on the pentacene layer through a shadow mask to form drain and source electrodes. The channel length L and width W are 30 μm and 200 μm respectively. Before the gold

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evaporation, the vacuum in the chamber was about 8x10^{-6} Torr.

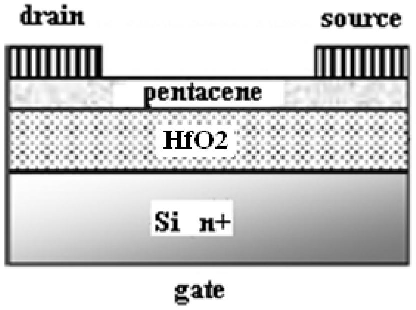

Fig. 1. Cross-sectional view of top-contact OTFT

The devices were characterized by HP4145B Semiconductor Parameter Analyzer, Berkeley Technology Associates FET Noise Analyzer Model 9603 and HP 35665A Dynamic Signal Analyzer to measure the I-V curve and 1/f noise characteristics of the organic thin-film transistors. The measurements were taken by a probe station in the ambient atmosphere.

III. RESULTS AND DISCUSSION

Fig. 2 and Fig. 3 reveal the output characteristics of the OTFTs with HfO_2 as the gate dielectric annealed in N_2O and NH_3 respectively. The OTFTs can work at very low operating voltage (less than 3.0 V), which is much lower than that of OTFT with SiO_2 as gate dielectric. The gate leakage is low when the former works at low drain-source voltage. Fig. 4 displays the comparison of the transfer characteristic of the two transistors. The on/off current ratio is about 4.3 ×10^3 for the former and 1.5x10^4 for the latter. Carrier mobility μ and threshold voltage V_{th} are two of the most important parameters to evaluate the performance of OTFTs. To deduce their values, the transfer characteristics curve of the OTFTs is plotted as $\sqrt{-I_d} = f(V_g)$, as shown in Fig. 4. According to the standard formula when OTFT operates in the saturation regime

$$I_d = -\frac{W}{2L}\mu C_{ox}(V_g - V_{th})^2 \tag{1}$$

we can get

$$\sqrt{-I_d} = \sqrt{\frac{W}{2L}\mu C_{ox}}(V_g - V_{th}) \tag{2}$$

where W and L are the width and the length of the channel respectively; C_{ox} is the oxide capacitance per unit area. From (2),

$$\mu = \frac{2L}{WC_{ox}}\left(\frac{\partial\sqrt{-I_d}}{\partial V_g}\right)^2 \tag{3}$$

After calculation, the carrier mobility is 0.399 cm^2/Vs for the N_2O-annealed device and 0.655 cm^2/Vs for the NH_3-annealed one. By extrapolating the $\sqrt{-I_d} = f(V_g)$ curve to the X-axis, the threshold voltage can be calculated. Besides, sub-threshold swing is also of great significance to analyze the switching characteristics of OTFTs. By plotting the transfer characteristics in the semi-logarithmic representation as in the inset of Fig. 4, the sub-threshold swing (SS) is found to be 0.361 V/dec and 0.229 V/dec for the N_2O-annealed and NH_3-annealed OTFTs respectively.

$$SS = \frac{1}{\frac{\partial Log(-I_d)}{\partial V_g}} \tag{4}$$

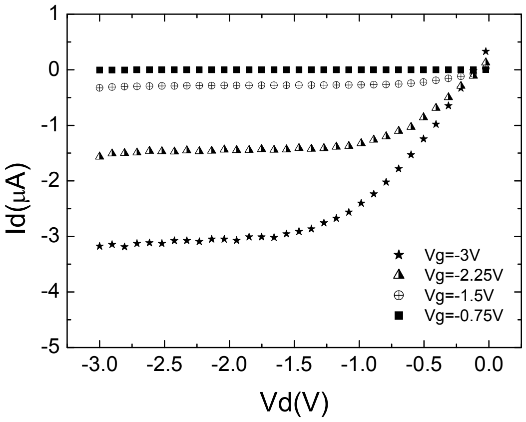

Fig. 2. Output characteristic of the OTFT annealed in N_2O

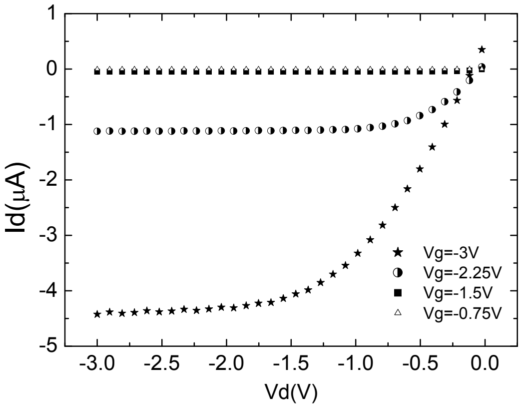

Fig. 3. Output characteristic of the OTFT annealed in NH_3

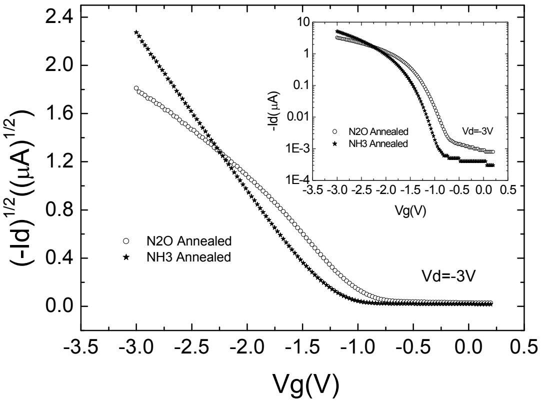

Fig. 4. Transfer characteristic of the OTFTs annealed in N2O or NH3 (Inset shows the Id-Vg curve in semi-logarithmic scale)

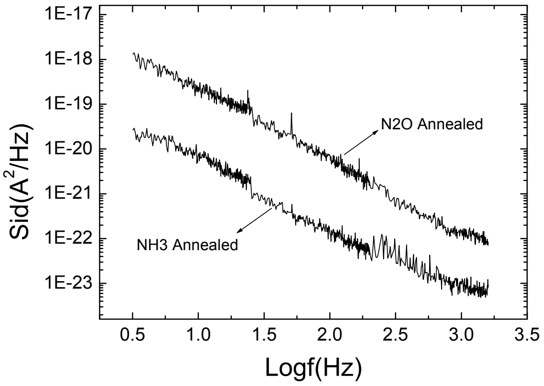

Fig. 5. 1/f noise characteristics of the OTFTs annealed in N_2O or NH_3

To further study the interface quality of the organic thin-film transistors, their 1/f noise spectrum was measured. The 1/f noise was tested in the frequency (*f*) range of 3.125 Hz and 1.6 kHz. Fig. 5 shows the comparison of the 1/f noise for the two transistors. The noise level of the transistor annealed in the NH_3 ambient is about 2 orders lower than that of its counterpart annealed in the N_2O ambient. It reveals that the device gain better interfacial quality through the NH_3 annealing than the N_2O annealing. In order to analyze the 1/f noise characteristic further, the Hooge parameter α is extracted according to the Hooge's empirical formula for the 1/f noise [9]

$$\frac{S_{id}(f)}{I_d^2} = \frac{\alpha}{Nf} \tag{5}$$

where S_{id} is spectral density of the 1/f noise of drain current I_d; and N is the total number of carriers in the channel. For the case of organic thin-film transistors, (5) can be approximated as [10],

$$\alpha = \frac{fS_{id}(f)L^2}{e\mu V_{ds} I_d} \tag{6}$$

where e is the electron charge. In this work, $f = 30Hz$ is used for the calculation of α. In the case of transistor which experienced the N_2O annealing at 200 °C, the Hooge parameter is 50.1. By contrast, the Hooge parameter is 0.274 for the transistor annealed in NH_3 at 200 °C. This means that the current fluctuation due to the 1/f noise in the former is more severe than the latter. In terms of sub-threshold swing, the transistor annealed in NH_3 is 0.229, which is better than 0.361 of the transistor annealed in N_2O. As a result, organic thin-film transistor annealed in NH_3 shows better switching characteristics than its counterpart annealed in N_2O. Besides, the former has higher carrier mobility than the latter. All these advantages of the transistor annealed in NH_3 over its counterpart annealed in N_2O can be attributed to its more nitrogen incorporation at the dielectric surface, and hence better interface quality between the gate dielectric and organic semiconductor. In other words, higher carrier mobility means weaker trap-related scattering for the carriers. Smaller 1/f noise and sub-threshold swing mean less traps in the channel / at the interface. On the other hand, the transistor annealed in NH_3 has a larger threshold voltage than the transistor annealed in N_2O, possibly because more nitrogen incorporation results in more positive oxide charges in the former. More quantitative analysis lies in better understanding of the carrier transport mechanism in the interface between the organic layer and the high-k dielectric layer of the organic thin-film transistors.

TABLE I

Device parameters of the OTFTs annealed in N_2O or NH_3

	Annealing gas	
	N_2O	NH_3
$C_{ox}(\mu F/cm^2)$	0.458	0.551
$t_{ox}(nm)$	11.2	10.9
k	5.8	6.8
$\mu(cm^2/Vs)$	0.399	0.655
$V_{th}(V)$	-0.801	-1.26
SS (V/decade)	0.361	0.229
on/off ratio (10^4)	0.432	1.52
$Id\|_{Vd=Vg=-3V}$ (μA)	3.17	4.42
α	50.1	0.274

IV. CONCLUSION

The application of HfO_2 as gate dielectric to pentacene thin-film transistors is studied in this paper. OTFTs with HfO_2 as gate dielectric were realized by RF sputtering and subsequent annealing in N_2O or NH_3 at 200 °C. Both can operate under a supply voltage of as low as 3 V. The OTFT annealed in NH_3 displays higher carrier mobility, larger on/off current ratio, smaller Hooge parameter and smaller sub-threshold swing than its counterpart annealed in N_2O. All these advantages make the organic thin-film transistor suitable in low-voltage and low-power applications. To conclude, HfO_2 annealed in NH_3 is very promising to act as the gate dielectric of high-performance OTFTs.

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