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Effects of fluorine plasma and ammonia annealing on pentacene thin-film transistor with ZrLaOx as gate dielectric

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Abstract
Pentacene organic thin-film transistor (OTFT) based on ZrLaOx gate dielectric is proposed and has been fabricated. The effects of fluorine plasma and ammonia annealing on the properties of the OTFT have been studied. It reveals that the plasma treatment can greatly improve carrier mobility and shift the threshold voltage in the positive direction. With a threshold voltage less than 0.5 V, the OTFT can work at very low supply voltage. On the other hand, the ensuing ammonia annealing counteracts the plasma treatment and shifts the threshold voltage in the opposite direction.

1. Introduction
Organic thin-film transistor (OTFT) is among the most promising candidates for future’s flexible displays, large-area sensor and radio-frequency identification tag [1-3]. Low cost, high throughput and lithography-free manufacturing are the advantages of the OTFT.

The operation and performance of OTFT highly depend on the interface between its gate dielectric and organic material. Both plasma treatment and annealing are efficient and convenient to improve the surface characteristics of the gate dielectric. Oxygen plasma [4] and N₂O plasma [5] have been reported to modify the dielectric surface and good results were obtained. Fluorine-based plasma has long been investigated to provide valuable surface properties in organic and inorganic material field [6]. Annealing was also used to modify the surface, and improvement of the OTFTs on the annealed HfLaO or SiO₂ as gate dielectric was reported [7, 8].

Generally, the operating voltage of OTFT based on SiO₂ is very large and reducing the thickness of gate dielectric inevitably leads to large leakage current. Therefore, using high-κ dielectric as the insulator layer is an alternative way to lower the threshold voltage and thin the device to make it more flexible [9].

In this study, high-κ ZrLaO₃ is proposed as the gate dielectric of pentacene OTFT and is prepared by sputtering. Also, the effects of fluorine plasma treatment and NH₃ annealing have been studied. With appropriate plasma treatment time, the OTFT can achieve high performance, e.g. small threshold voltage as well as high carrier mobility.

2. Device fabrication
Pentacene OTFTs with ZrLaO₃ gate dielectric were fabricated by employing the bottom-gate top-contact configuration. Heavily doped silicon wafers (n-type, <100>, resistivity of 0.5 ~ 0.7 Ω⋅cm) were cleaned according to the standard RCA method and dipped in dilute HF acid (2%) to remove the native oxide. Next, ZrLaOₓ dielectric was sputtered at room temperature by a radio-frequency sputterer (Denton Vacuum LLC Discovery 635). After that, eight wafers were divided into four groups, which then were treated by a fluorine plasma, with CHF₃ at 10 sccm and O₂ at 1 sccm for different times (0 s, 100 s, 300 s and 600 s). Then, for each group, one wafer was annealed at 400 °C in NH₃, at a flow rate of 1000 mL/min for 10 min. Next, 30-nm pentacene (purchased from Sigma-Aldrich without purification) was evaporated on the dielectric by an evaporator (Edwards Auto 306). The film was evaporated in high vacuum (5×10⁻⁶ torr) on the wafers at a deposition rate of 1.2 nm/min, as monitored by the quartz-crystal oscillator. Lastly, the drain and source electrodes were formed on the pentacene film by gold evaporation through a shadow mask. The width and length of the channel on the shadow mask were 200µm and 30 µm, respectively.

The I-V characteristics of the transistors were measured by HP 4145B Semiconductor Parameter Analyzer, Berkeley Technology HP 3565A Dynamic Signal Analyzer. Al/ZrLaOₓ/heavily-doped Si structure was used to fabricate capacitors by lithography for measuring the dielectric capacitance which was measured by HP 4284A Precision LCR Meter. All the
The field-effect carrier mobility of the OTFTs is extracted by the MOSFET model. In the saturation regime:

\[ I_D = \left( \frac{W}{2L} \right) C_{ox} \mu (V_G - V_T)^2 \]  

(1)

where \( W \) is the channel width, \( L \) the channel length, \( C_{ox} \) the insulator capacitance per unit area, \( I_D \) the saturation drain-source current, \( V_G \) the gate voltage and \( V_T \) the threshold voltage.

From equation (1), the field effect mobility is expressed by

\[ \mu = \frac{2L}{WC_{ox}} \left( \frac{\partial \sqrt{-I_D}}{\partial V_G} \right)^2 \]  

(2)

The sub-threshold swing (SS) of the OTFTs is usually used to measure the speed that the device switches between the "off" state and the "on" state. It could be calculated by the equation (3).

\[ SS = \frac{1}{\frac{\partial \log_{10}|I_D|}{\partial V_G}} \]  

(3)

Figure 1 and Figure 2 show the transfer characteristics of the OTFTs. The main parameters extracted from them are listed in Table 1 and Table 2. The plasma treatment can greatly improve the carrier mobility. The \( \text{NH}_3 \) annealing seems to have little effect on the carrier mobility improvement when the dielectric of the OTFTs is not treated by the plasma, which is different from the reports [10, 11]. However, the annealing after the plasma treatment can compensate the effects of the plasma on the carrier mobility as shown in Tables 1 and 2. With a plasma treatment time of 300s, the carrier mobility of both groups reaches its highest value, 0.417 cm²/Vs and 0.178 cm²/Vs, respectively. After that, the carrier mobility begins to decrease with the plasma treatment time. This is mainly because excessive plasma treatment can cause damage and traps at the dielectric surface, and thus increase the carrier scattering, resulting in lower carrier mobility [12].
surface, and thus shift the threshold voltage in the opposite direction. In summary, all the transistors can work at very low supply voltage because high-κ ZrLaO\textsubscript{x} dielectric is used. Moreover, the threshold voltage can be adjusted by the plasma treatment or the NH\textsubscript{3} annealing.

The sub-threshold swing also increases after the annealing as shown in Tables 1 and 2. Since the maximum current increases after the plasma treatment, the on/off ratio is also improved.

The OTFT with 600-s plasma transistors can operate with a low supply voltage of less than 6 V. Among them, the OTFT with 600-s plasma-treated OTFTs without annealing and with treatment achieves the largest drive current of above 5 \(\mu\text{A}\), while the OTFT without plasma treatment and with the annealing has the smallest drive current of about 0.4 \(\mu\text{A}\). Although the maximum currents of the transfer characteristics for the 100-s, 300-s, and 600-s plasma-treated OTFTs without annealing are similar, the maximum current of the 300-s plasma-treated sample is much greater than those of the others as shown in Figure 3.

4. Summary

The effects of fluorine plasma treatment and NH\textsubscript{3} annealing on the ZrLaO\textsubscript{x} gate dielectric of pentacene OTFT have been investigated and the carrier mobility of the device can be greatly improved by the plasma treatment. The plasma treatment can increase the density of charge traps, and thus shift the threshold voltage in the positive direction. On the contrary, the annealing can passivate the traps and cause the threshold voltage to have a negative shift. Therefore, through the use of the plasma treatment or the annealing, the threshold voltage of the OTFT can be properly adjusted to suit the applications.

References