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

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Abstract—Pentacene organic thin-film transistor (OTFT) with high- κ HfTiO gate dielectric has been fabricated. The effects of fluorine plasma and ammonia annealing on the properties of the OTFT have been studied. After treating the dielectric in the plasma, the carrier mobility of the transistor can be improved by about 5 times to $0.0883 \text{ cm}^2/\text{V}\cdot\text{s}$. Moreover, the fluorine plasma treatment can shift the threshold voltage of the device in the positive direction. Experimental results also show that NH_3 annealing can enhance the OTFT performance in terms of higher mobility, smaller sub-threshold slope and larger on/off ratio.

Keywords—OTFT; high κ ; HfTiO; dielectric

I. INTRODUCTION

Organic thin-film transistors (OTFTs) employing various organic semiconductor materials were extensively investigated over the last four decades due to their potential applications in sensors, flat-panel displays and electronic identification tags [1, 2]. Many organic semiconductors have been used to make OTFTs, e.g. pentacene, rubrene, perylene, poly(3-hexylthiophene) regioregular (P3HT) and dihexylquin-quethiophene (DH-5T). Among these organic materials, pentacene is commonly chosen as active layer for making OTFT. It is because pentacene is a strong molecular-crystal former, which is an important factor in obtaining high field-effect mobility [3]. Moreover, pentacene is a p-type semiconductor, which is stable in air and less sensitive to moisture. Silicon dioxide and oxynitride have traditionally been used as the gate insulator in OTFTs. However, these kinds of insulators are not very suitable for making high-performance OTFTs for high-speed and low-power display driving circuits. In this study, sputter-deposited high- κ material hafnium titanium oxide HfTiO is used as the gate dielectric for the pentacene OTFTs. High- κ materials can allow thicker physical thickness to suppress gate leakage current while maintaining a high gate capacitance to reduce operating voltage of OTFTs. The gate dielectric is then annealed in N_2 or NH_3 , followed by fluorine plasma treatment for various durations to achieve better quality. The electrical characteristics of the OTFTs are studied and compared.

II. RESULTS AND DISCUSSION

Fig. 1 shows the electrical output characteristics of the OTFTs with gate dielectric annealed in N_2 or NH_3 at 400°C followed by plasma treatment for 0, 100, 600 or 900 s. All of the OTFTs have very low operating voltages and show good saturation behavior. It is found that the drain current increases with plasma treatment time for both N_2 and NH_3 -annealed samples.

The transfer characteristics of the OTFTs are shown in Fig. 2. The field-effect mobility μ can be calculated from the slope of the plot of $(-I_D)^{1/2}$ versus V_G in the saturation regime. The sub-threshold slope (SS) is calculated by taking the reciprocal of the steepest slope of the logarithmic plot of I_D versus V_G . This is a key parameter for OTFTs used as switches because it shows the voltage needed to distinguish between the on and off modes of the switch. The significant parameters of the devices are summarized in Tables I and II. Without fluorination, the carrier mobility of the OTFTs annealed in NH_3 is $\sim 40\%$ higher than that of its counterpart with N_2 annealed. It is because during ammonia annealing, atomic N and H decomposed from NH_3 can passivate the surface of the HfTiO film and decrease the traps at the surface. The trap-related carrier scattering is thus reduced, resulting in higher carrier mobility [4]. The sub-threshold slope and on/off ratio of the OTFTs with NH_3 -annealed HfTiO as gate dielectric are 22% smaller and 69% larger than those of the OTFTs with N_2 -annealed HfTiO respectively. These enhanced electrical characteristics of the NH_3 annealed sample are attributed to the nitridation-induced passivation of the dielectric surface, which can reduce the traps in the channel of the OTFT to produce a better insulator/organic interface. From the sub-threshold slope, the density of surface states at the semiconductor/dielectric surface of the NH_3 -annealed OTFT is $1.84 \times 10^{13} \text{ cm}^{-2}$, which is 21% smaller than that of the N_2 -annealed sample.

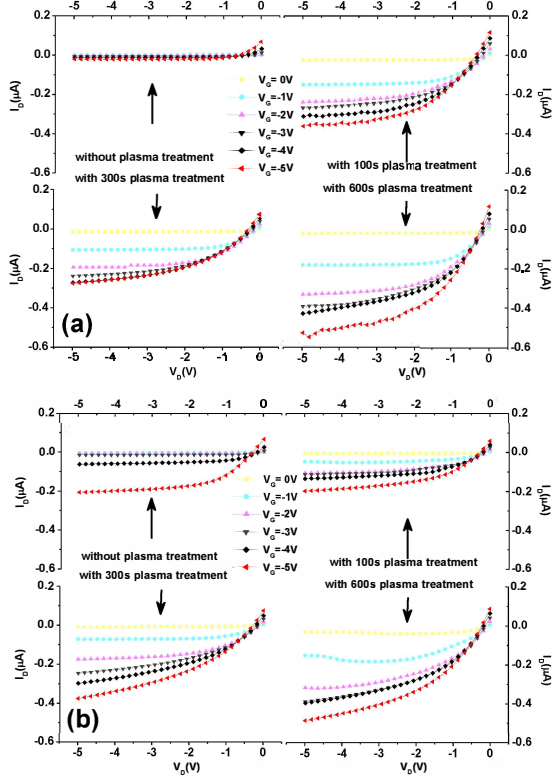
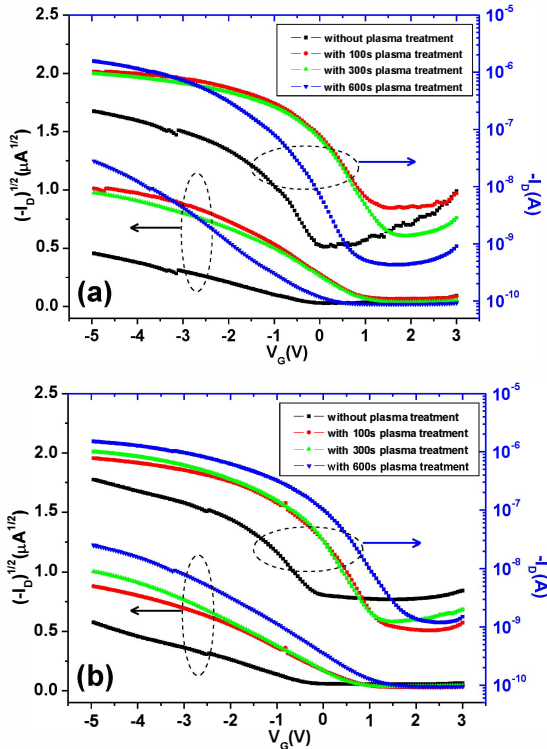
Moreover, the fluorine plasma treatment can improve the carrier mobility. The carrier mobility of the device is found to increase with the plasma treatment time because more fluorine atoms can incorporate at the HfTiO surface, resulting in less dangling bonds at the gate-dielectric surface (due to the formation of strong Hf-F bonds) and less oxygen vacancies [5], thus less trap-related carrier scattering. Compared to the control samples without fluorine incorporation, both the N_2 and NH_3 -annealed OTFTs with 100-s plasma treatment have higher mobility, smaller sub-threshold and larger on/off ratio. These improvements should be due to the passivation effects of F. Lastly, the threshold voltage of all the OTFTs shifts toward the positive direction after fluorination. This should be due to negative fluorine ions occupying the interstitial sites of the gate dielectric.

TABLE I. PARAMETERS OF OTFTS ANNEALED IN N₂ AT 400°C

Sample No.	A	B	C	D
Plasma Treatment Time (s)	0	100	600	900
Mobility (cm ² /V·s)	0.0170	0.0534	0.0648	0.0883
Threshold Voltage (V)	-0.452	0.824	1.05	0.712
SS (V/dec)	0.898	0.777	0.750	0.580
On/Off Current Ratio (10 ³)	0.244	0.340	0.685	1.54
I _D (V _{DS} = V _{GS} = -5 V) (μA)	0.183	0.999	1.10	1.41
C _{ox} (μF/cm ²)	0.267	0.317	0.296	0.283
t _{ox} (nm)	47.5	47.5	47.2	47.0
k	14.3	16.9	15.8	15.0

 TABLE II. PARAMETERS OF OTFTS ANNEALED IN NH₃ AT 400°C

Sample No.	E	F	G	H
Plasma Treatment Time (s)	0	100	600	900
Mobility (cm ² /V·s)	0.0240	0.0405	0.0459	0.0631
Threshold Voltage (V)	-0.554	0.361	0.826	1.25
SS (V/dec)	0.702	0.687	0.771	0.821
On/Off Current Ratio (10 ³)	0.412	0.708	0.818	0.349
I _D (V _{DS} = V _{GS} = -5 V) (μA)	0.256	0.631	0.904	1.29
C _{ox} (μF/cm ²)	0.273	0.270	0.312	0.303
t _{ox} (nm)	48.1	47.4	47.2	47.1
k	14.9	14.5	16.7	16.1


 Fig.1. Output characteristics of the OTFTs: (a) annealed in N₂; (b) annealed in NH₃.

 Fig.2. Transfer characteristics of the OTFTs: (a) annealed in N₂; (b) annealed in NH₃.

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