

Study on the Hydrogen Treatment Effect of Vacuum deposited Pentacene Thin Film Transistors

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Abstract

In order to reach the high electrical quality of organic thin film transistors (OTFTs) such as high mobility and on-off current ratio, it is strongly desirable to study the enhancement of electrical properties in OTFTs. Here, we report the novel method of hydrogen (H₂) plasma treatment to improve electrical properties in inverted staggered OTFTs based on pentacene as active layer. To certify the effect of this method, we compared the electrical properties of normal device as a reference with those of device using the novel method. In result, the normal device as a reference making no use of this method exhibited a field effect mobility of 0.055 cm²/Vs, on/off current ratio of 10³, threshold voltage of -4.5 V, and subthreshold slope of 7.6 V/dec. While the device using the novel method exhibited a field effect mobility of 0.174 cm²/Vs, on/off current ratio of 10⁶, threshold voltage of -0.5 V, and subthreshold slope of 1.49 V/dec. According to these results, we have found the electrical performances in inverted staggered pentacene TFT owing to this novel method are remarkably enhanced. So, this method plays a key role in highly improving the electric performance of OTFTs. Moreover, this method is the first time yet reported for any OTFTs

1. Introduction

Organic thin film transistors (OTFTs) based on pentacene as active layer have recently received considerable interest. OTFTs offer advantages compared to traditional field-effect transistors, like mechanical flexibility and weight reduction. [1-2] But because of the relatively low mobility and on-off current ratio of OTFTs, they cannot rival the performance of field-effect transistors based on single crystalline inorganic semiconductors, such as Si, Ge, and GaAs, which have charge carrier mobility of three or more orders of magnitude higher. [3] To make OTFTs with high mobility and on-off current ratio, it is very important to enhance the electrical properties

of organic thin film based pentacene itself. In detail, the interface defects, traps, and crystallinity of pentacene are crucial parameters that can influence electrical properties of OTFTs based pentacene.

In this work, in order to improve the electrical properties of pentacene TFT, we have studied the hydrogen plasma treatment effect on inverted staggered pentacene TFT. We used pentacene as an active layer, silicon-oxide (SiO₂) as a gate insulator, Cr as a gate electrode, and Au as source/drain electrodes. We compared device using hydrogen plasma treatment technique with device without hydrogen plasma treatment technique. And then, we analyzed the pentacene film and the electrical properties of inverted staggered pentacene TFT by means of Secondary Ion Mass Spectroscopy (SIMS), Atomic Forced microscopy (AFM) image. In view of the our results so far achieved, our experimental data will show the electrical performance of inverted staggered pentacene TFT is strongly dependent on that the change of the morphology and structure of pentacene thin film using hydrogen plasma treatment. Moreover, according to our experimental data, the off-current and threshold voltage of inverted staggered pentacene TFT are remarkably reduced by hydrogen plasma treatment on pentacene. On the other hand, the on current and field effect mobility of pentacene TFT are highly enhanced by hydrogen plasma treatment.

2. Experiment

The fabrication process of the inverted staggered pentacene TFT using the novel method of hydrogen plasma treatment was as follows; First, a 70 nm thick Chromium layer was deposited by sputtering. And then this layer was patterned as a gate electrode

using photolithography. Undefined chromium was etched away in a ($\text{HClO}_4 + \text{NH}_4\text{CeNO}_3$) solution. On the patterned gate electrode, a 150nm thick silicon oxide (SiO_2) as a gate insulator was deposited by plasma enhanced chemical vapor deposition (PECVD) at a substrate temperature of 250°C and RF power of 10 W. The flow rates of He, SiH_4 , N_2O were fixed at 200, 150, 400 sccm, respectively. After patterning the gate insulator using photolithography, undefined SiO_2 was etched away in a ($\text{NH}_4\text{F} + \text{HF} + \text{H}_2\text{O}$) solution.

Second, On gate insulator, a 150nm thick pentacene films as an active layer was deposited by a thermal evaporation in a high-vacuum environment with typical background pressure of 10^{-6} torr at a substrate temperature of 80°C . In this paper, as-received pentacene is used without additional purification. The power of pentacene is loaded in alumina crucible that is resistively heated. The temperature of the sample holder can be controlled in a large temperature range by resistive heating elements and water-cooling. The temperatures of the substrate and the crucible are monitored by chromel-alumel thermocouples. An Intelmetrix IL 400 quartz oscillator monitors the deposition rate. A shutter allows adjusting the evaporation rate to the desired value (within an accuracy of 0.1 nm/s) before the actual deposition of the thin film. After evaporation the substrate is slowly cooled (rate $\sim 0.7^\circ\text{C}$ per second) to room temperature in order to prevent the damage of pentacene film due to the sudden phase transition.[4-5] Active layer was patterned using shadow mask. After then, with increasing the time of hydrogen plasma treatment from 30 sec to 90sec, the H_2 plasma treatment was carried out on the pentacene film as an active layer in a PECVD reactor at RF power of 40 W. The flow rate of H_2 was fixed at 10 sccm (figure 1). After H_2 plasma treatment, a 100nm thick gold layer as a source/drain electrode was deposited by sputtering. During deposition of gold layer, source/drain electrode was defined using shadow mask.

3 Result and Discussion

Figure 2 (a) and (b) show drain current gate voltage (I_D - V_G) characteristics for the inverted staggered pentacene TFTs in the comparison between

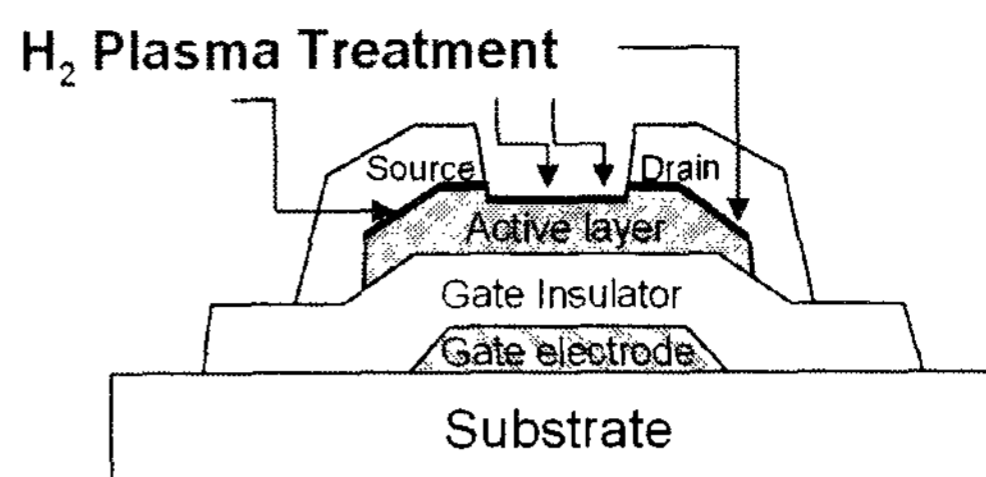


Figure 1. Schematics of the inverted staggered pentacene TFTs showing H_2 plasma treatment on pentacene layer.

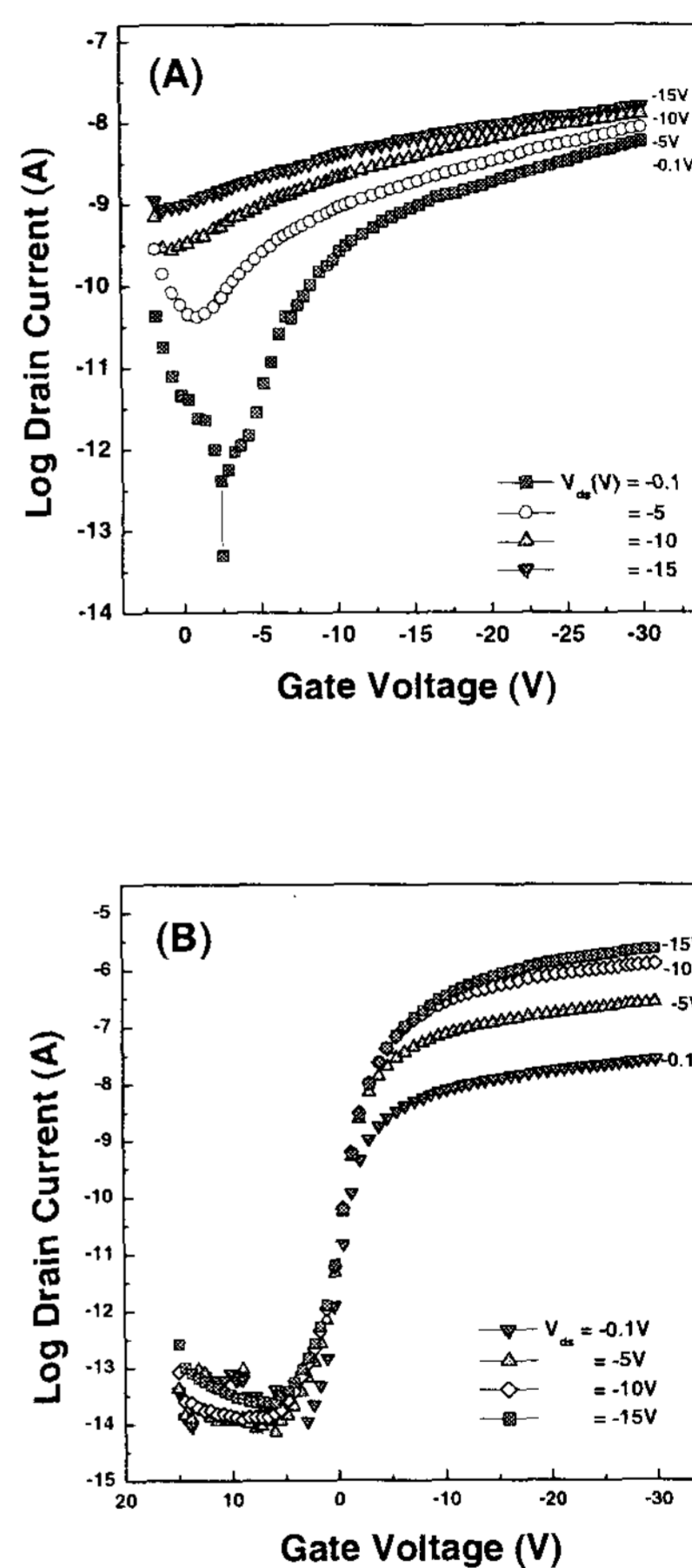


Figure 2. Drain current gate voltage (I_D - V_G) characteristics for the inverted staggered pentacene TFTs (a) without H_2 plasma treatment (b) with H_2 plasma treatment during the plasma time of 90 sec. ($W/L=5000/2000$)

without H_2 plasma treatment and with H_2 plasma treatment. For the device without H_2 plasma treatment (figure 3 (a)), the electrical properties exhibited an off current of 10^{-11} A, on current of 10^{-8} A, and

subthreshold slope of 7.6 V/dec at $V_D = -0.1$ V. While the device with H_2 plasma treatment (figure 3 (b)) exhibited an off current of 10^{-11} A, on current of 10^{-7} A, and subthreshold slope of 1.49 V/dec at $V_D = -0.1$ V. the on/off current ratio of the inverted staggered pentacene TFTs with H_2 plasma treatment are greater than that of the inverted staggered pentacene TFTs without H_2 plasma treatment. These results demonstrate that the electrical properties are enhanced due to the effect of hydrogen plasma treatment. That is, RMS (root-mean-square) roughness and trap state due to grain boundaries at pentacene layer were reduced during H_2 plasma treatment. Also the contact resistance between pentacene and source/drain electrode was reduced.

Figure 4 (a) and (b) show the field effect mobility and threshold voltage for the pentacene TFTs in the comparison between without hydrogen plasma treatment and with hydrogen plasma treatment. They were obtained in the saturation region using the following relation:

$$I_D = (W/2L) C_i \mu_n (V_G - V_{TH})^2; \quad (1)$$

Where W is the channel width, L is channel length of the TFT and C_i is the capacitance of the gate insulator. Equation (1) is conventionally used to characterize OTFTs.

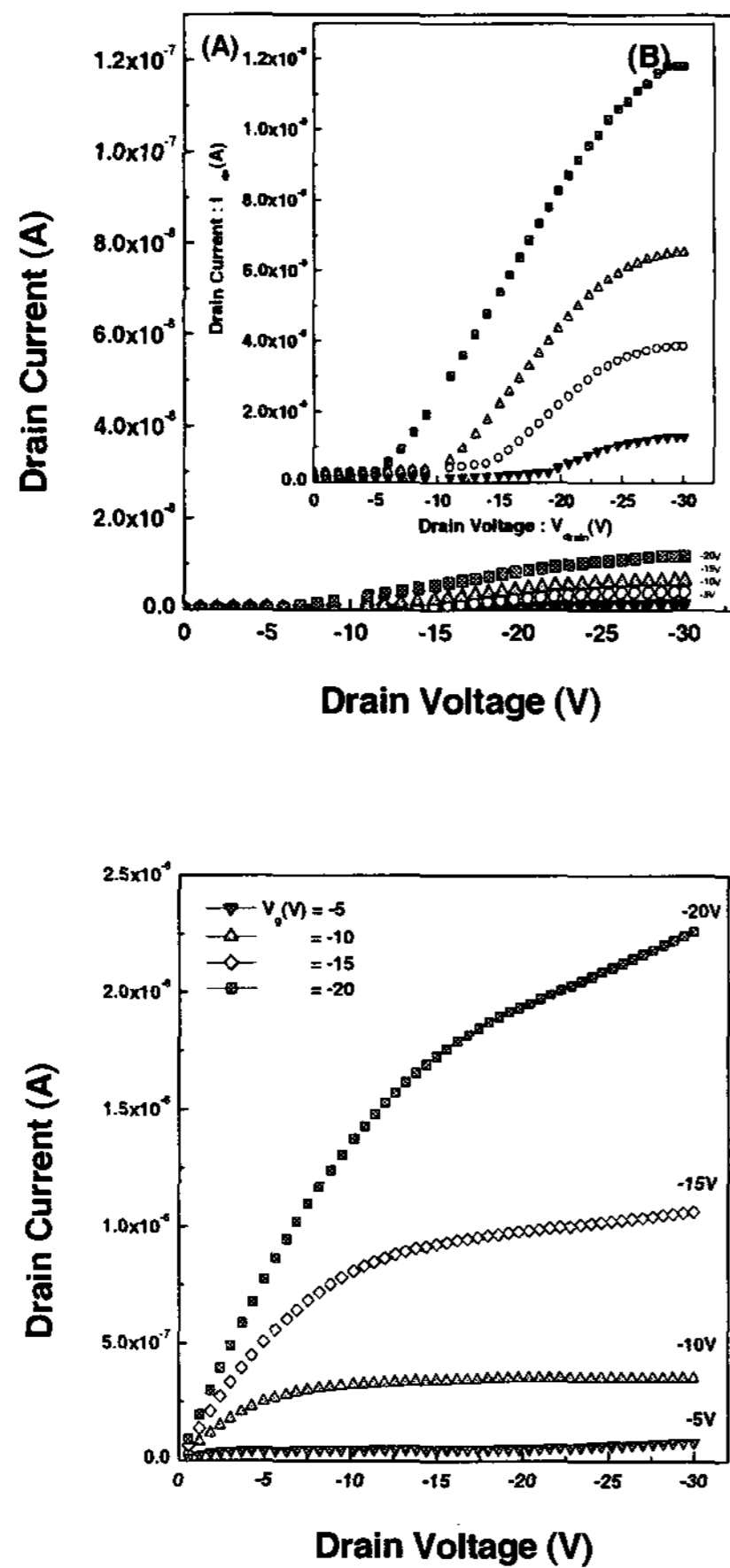


Figure 3. The output characteristics of the inverted staggered pentacene TFTs (a) without H_2 plasma treatment (b) with H_2 plasma treatment during the plasma time of 90 sec. ($W/L=5000/2000$)

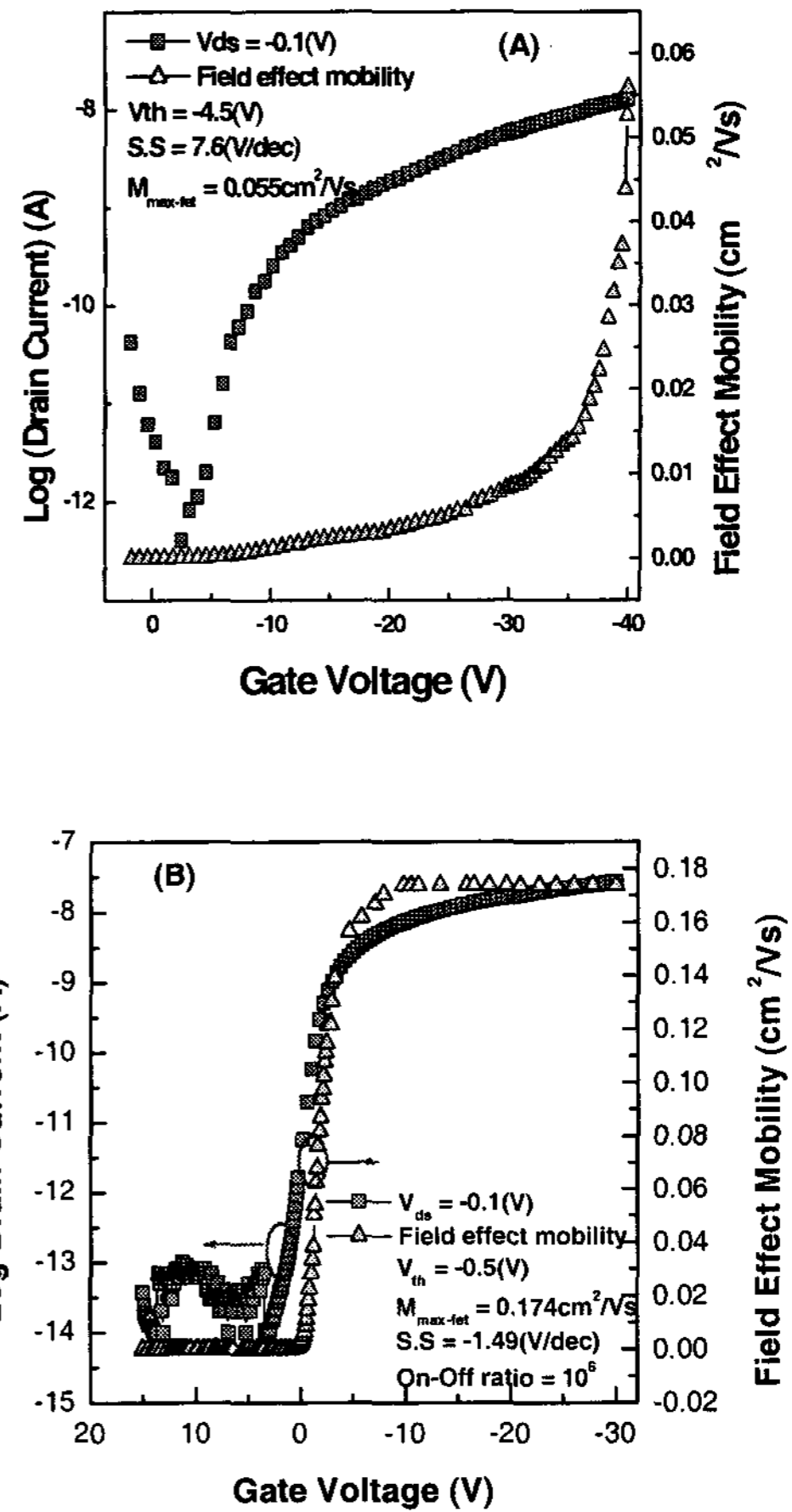


Figure 4. The field effect mobility and threshold voltage for the pentacene TFTs (a) without H_2 plasma treatment (b) with H_2 plasma treatment during the plasma time of 90 sec. ($W/L=5000/2000$)

The device without H₂ plasma treatment (figure 4 (a)) exhibited a field effect mobility of 0.055 cm²/Vs and threshold voltage of -4.5 V at V_D = -0.1 V. While the device with H₂ plasma treatment (figure 4 (b)) exhibited field effect mobility of 0.174 cm²/Vs and threshold voltage of -0.5 V at V_D = -0.1 V. This data also show that the field effect mobility of the pentacene TFTs with hydrogen plasma treatment is greater than that of without hydrogen plasma treatment about 2 orders of magnitude. According to our expectation, charging effect on pentacene layer due to H₂ plasma treatment is reduced and the hydrogen is passivated in the defect of pentacene layer. In the result, the field effect mobility and threshold voltage is highly enhanced.

Figure 5 (a) shows the thickness of pentacene films plotted against hydrogen plasma treatment time. We measured the thickness of pentacene film after the hydrogen plasma treatment. The thickness of pentacene film decreases slightly with increasing hydrogen plasma treatment time. This is due to the surface etching or to the structural relaxation of pentacene film by hydrogen plasma. Figure 5 (b) and (c) shows hydrogen plasma treatment time dependence of rms roughness of the hydrogen plasma treated pentacene surfaces obtained from atomic force microscopy (AFM) images. The RMS roughness of pentacene surface without hydrogen plasma treatment is 16.8 nm. But the RMS roughness of pentacene film with H₂ plasma treatment is decreased to 6nm. According to our expectation, the pentacene surface can be modified by a reaction between the atoms at the surface and hydrogen metastables. The surface atoms can be relaxed by the energy transferred from the conversion of metastable hydrogen atoms to ground state. Also H₂ plasma treatment plays a role in removing particle of an occasion for degradation on pentacene and reduction of grain boundaries in pentacene. This effects result in enhancement of electrical properties such as field effect mobility.

Figure 6 shows SIMS (a) hydrogen, (b) oxygen, and (c) C-H bond depth profiles in pentacene layer with H₂ plasma treatment. In the case of SIMS hydrogen depth profiles, during plasma treatment time of 30 seconds, the H₂ atom is reduced. During this period, the ashing on pentacene layer took effect. But after that, the H₂ atom is increased and diffused on the pentacene layer. Because of this phenomenon, trap and grain boundaries were reduced. Therefore, conductivity in pentacene TFT is enhanced. In the

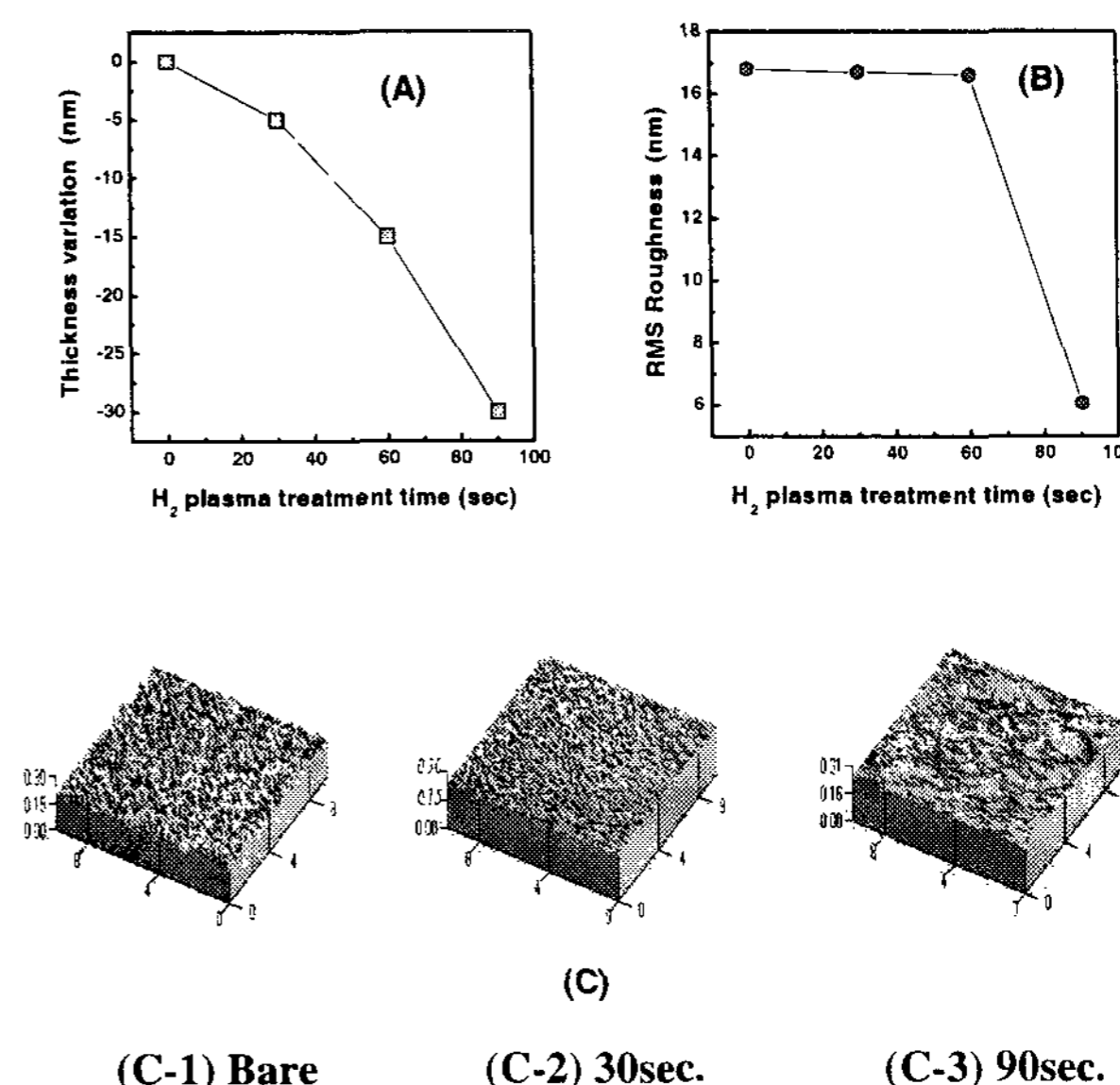


Figure 5. H₂ plasma treatment time dependence of (a) rms roughness (b) Thickness variation (c) AFM 3D image of the H₂ plasma treated pentacene surfaces obtained from atomic force microscopy (AFM).

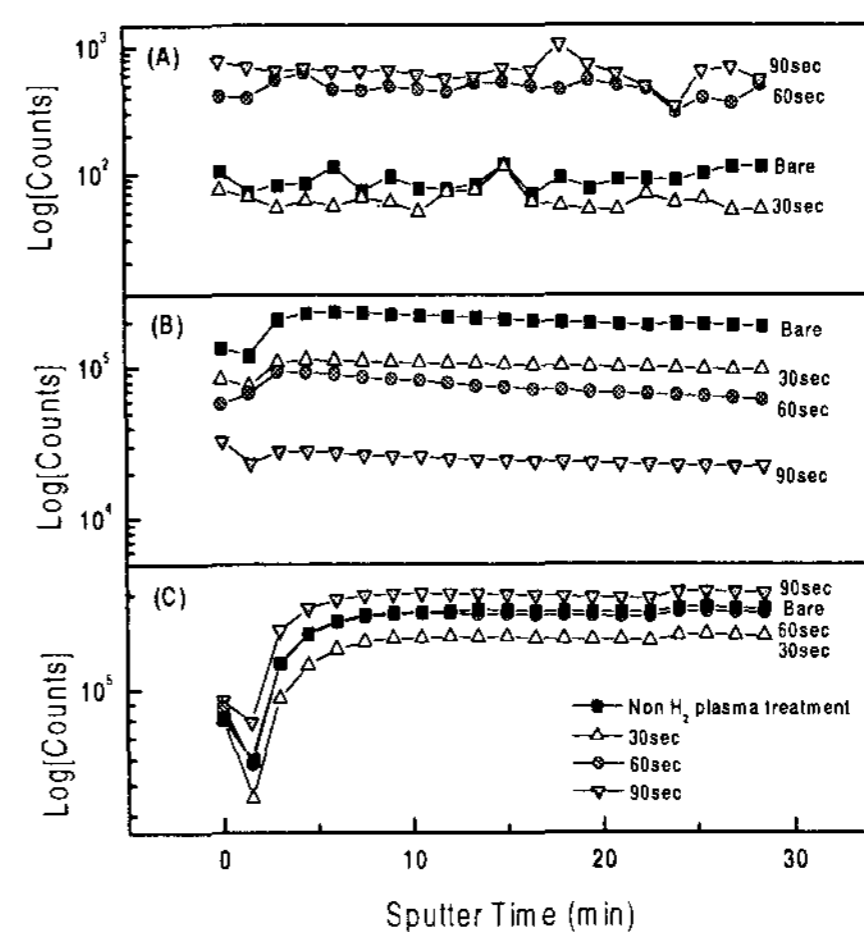


Figure 6. SIMS (a) hydrogen depth profiles, (b) oxygen depth profiles, and (c) C-H bond depth profiles in pentacene layer using H₂ plasma treatment.

case of SIMS O₂ depth profiles, with increasing H₂ plasma treatment time, O₂ atom is reduced. The cause of degradation in pentacene TFT is existing O₂ atom on pentacene layer. The reduction of O₂ atom on pentacene layer gave rise to enhancement of electrical performance in pentacene TFT. In the case of SIMS C-

H bond depth profiles in pentacene layer with H₂ plasma treatment, during plasma treatment time of 30 seconds, C-H bond is reduced due to occurrence of ashing effect. After that, this bond is increasingly recovered and increased. In this state, H₂ atom is passivated on pentacene layer. H₂ atom passivated on pentacene layer plays a role in enhancement of charging effect in pentacene layer itself. Therefore the electrical properties in pentacene TFT are highly improved.

4 Conclusion

Using this approach of the inverted staggered pentacene TFTs with H₂ plasma treatment on pentacene layer, we find that H₂ plasma treatment on pentacene layer strongly depends on the electrical performance of pentacene TFTs. Actually; we observe that the electrical performance of pentacene TFTs is enhanced by H₂ plasma treatment. This means that hydrogen plasma treatment on OTFTs play a key role in determining the high electric performance of organic thin film transistor devices. The fabricated inverted staggered pentacene TFTs using H₂ plasma treatment with a field effect mobility of 0.174 cm²/Vs, on/off current ratio of 10⁶, threshold voltage of -0.5 V, and subthreshold slope of 1.49 V/dec V_D = -0.1 V are the first time yet reported for any OTFTs.

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