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Jaehoon Park^a, Hyunsuck Kim^a, Jae-Hoon Kim^a,
Jong Won Lee^b & Jong Sun Choi^b

^a Department of Electronics and Computer Engineering, Hanyang University, Seoul, Korea

^b Department Electrical, Information and Control Engineering, Hongik University, Seoul, Korea

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Characteristics of Organic Thin-Film Transistors with Self-Assembled Monolayer Formed in Gas-Phase on PVP Gate Insulator

Jaehoon Park¹, Hyunsuck Kim¹, Jae-Hoon Kim¹,
Jong Won Lee², and Jong Sun Choi²

¹Department of Electronics and Computer Engineering,
Hangyang University, Seoul, Korea

²Department Electrical, Information and Control Engineering,
Hongik University, Seoul, Korea

The electrical characteristics of organic thin film transistors (OTFTs) with self-assembled monolayer (SAM) on the poly(4-vinylphenol) gate insulator have been investigated. The SAM was formed in gas-phase using the atomic layer deposition method onto gate insulator. It is observed that SAM modifies the surface polarity of gate insulator and influence the growth of the subsequent pentacene layer. Improvements of the crystallinity and directionality of pentacene molecules are confirmed by X-ray diffraction measurement. Also, the roughness of the pentacene thin film and the device performance are improved.

Keywords: ALD; organic thin-film transistor; pentacene; SAMs

INTRODUCTION

Semiconducting π -conjugated materials have received considerable attention as active components in electronic devices such as light emitting diodes, photovoltaic cells, field-effect transistors, and their integrated organic devices [1]. Organic materials can offer substantial advantages in terms of the processing simplicity and competitive cost [2]. The performance of these organic devices, especially organic thin-film transistor (OTFTs), has significantly improved during the past several years. The optimized OTFTs have shown the electrical

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Address correspondence to Dr. Jaehoon Park, 17 Haengdang-dong, Seongdong-gu, Hanyang University, IT 1210, Seoul 133-791, Korea. E-mail: jaypark0420@gmail.com

characteristics comparable to those obtained with amorphous silicon devices. It has been reported that pentacene ($C_{22}H_{14}$) shows the most pronounced properties for *p*-channel conduction, among many organic semiconductors. For this reason, pentacene is usually employed as the active layer of OTFTs. And, polymer gate dielectric materials, such as poly(4-vinylphenol) and poly(methyl-methacrylate), have been adopted for the OTFTs [3].

One of critical factors for high-performance OTFTs is the properties of the pentacene-gate dielectric interface. Pentacene exhibits a strong tendency to form highly ordered film depending on the growth and substrate surface condition [4]. It is known that the vertical alignment of pentacene molecules to the gate insulator surface provides a strong π - π^* overlap and increases the electrical conductivity in the direction of perpendicular to the long-axis. The hydrophilic surface of PVP gate insulator can deteriorate the upright growth of pentacene molecules to the substrate. Therefore, in order to align pentacene molecules upright onto the gate insulator surface with high order, certain surface treatments such as rubbing, photo-aligning, self-assembled monolayer (SAM) coating, are necessary, which can contribute to the characteristic improvements in OTFTs [5].

In this work, SAM was deposited onto the PVP layer prior to the deposition of pentacene in order to modify the polarity of insulator surface. Conventional coating processes dipping substrates into the SAM solution have some problems associated with processing complexity, uniformity, reproducibility and possibility to harm to polymeric gate insulator. Thus, as an alternative, we attempt to form SAM onto the PVP layer in gas-phase using atomic layer deposition (ALD) process. The properties of pentacene thin film and characteristics of OTFTs with the ALD-deposited SAM are presented.

EXPERIMENTAL DETAILS

The structure of the fabricated OTFTs is shown in Figure 1. In the fabrication of OTFTs, a 100-nm-thick Al layer was thermally evaporated through a shadow mask onto a glass substrate as a gate electrode. The PVP gate insulator was spin-coated to be 300 nm thick, which was confirmed by the α -step profilometer. For the formation of SAM, a 1,2-nm-thick octyltrichlorosilane ($CH_3(CH_2)_7SiCl_3$) monolayer was deposited onto the PVP-coated substrate using ALD method under the base pressure of 5×10^{-3} Torr. In detail, the PVP-coated substrate and the octyltrichlorosilane liquid were heated to 150°C and 170°C, respectively. And then, the octyltrichlorosilane vapor was carried by Ar (99.999%, 50 sccm) gas and adsorbed to the surface

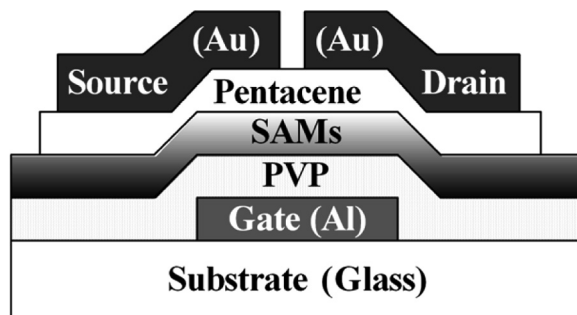


FIGURE 1 Structure of the fabricated OTFTs.

of PVP layer. The remnants of octyltrichlorosilane vapor were purged from the ALD chamber using Ar (99.999%) gas. And a 60-nm-thick pentacene layer was thermally evaporated under a pressure of 10^{-6} Torr with the deposition rate of 0.05 nm/s using the second shadow mask. Then a 50-nm-thick Au layer was thermally evaporated as source/drain electrodes using the third shadow mask. Devices with top-contact geometry were fabricated, in which the channel length (L) and width (W) are 90 μm and 300 μm , respectively.

RESULTS AND DISCUSSION

The change of the surface polarity of the SAM-treated PVP layer was confirmed by contact angle analysis as shown in Figure 2. In order to calculate the surface energy, water and diiodomethane were used. It is observed that SAM changes the surface of PVP layer more hydrophobic and decreases its surface energy, which means that the factor deteriorating the upright growth of pentacene molecules could be suppressed. Measured contact angles and computed surface properties are listed in Table 1.

The surface morphology of the pentacene thin film is observed by atomic force microscopy (AFM) as shown in Figure 3, where the height is adjusted to be 60 nm. It is observed that the grain size of the pentacene film on the SAM becomes uniform and the crevice between the grains gets shallow. And also, the average roughness of the pentacene film on the SAM-treated PVP layer (about 3.1 nm) is improved compared with that on the bare PVP (about 4.7 nm).

According to the AFM images, it is expected that the SAM affect the growth condition and the crystallinity of the pentacene film. In order to confirm the change of the crystallinity, x-ray diffraction (XRD)

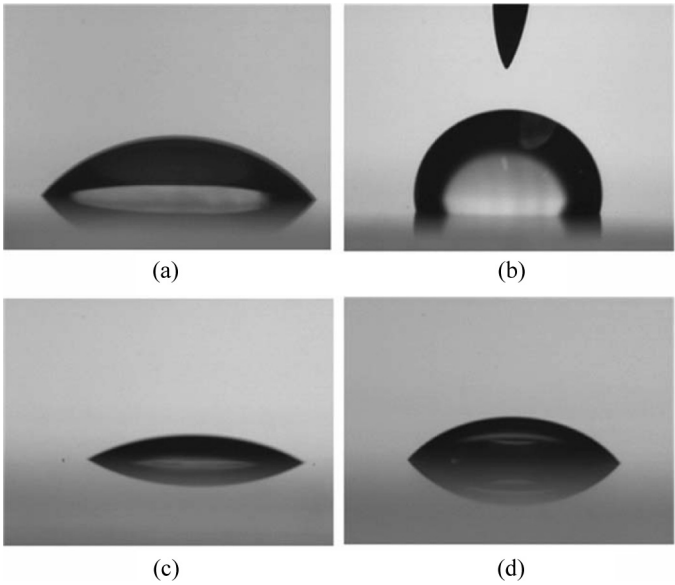


FIGURE 2 Contact angles of (a) water droplet on the bare PVP, (b) water droplet on the SAM-treated PVP, (c) diiodomethane droplet on the bare PVP and (d) diiodomethane droplet on the SAM-treated PVP.

measurement is carried out using 60-nm-thick pentacene layers on different substrates. XRD patterns of pentacene film on the PVP layer without and with SAM are shown in Figures 4(a) and (b), respectively. It is observed that the thin-film phase is intensive for both films but the thin-film phase for that on the SAM shows slightly upright pattern [6,7], meaning that the charge transport in this film can be improved owing to an efficient π orbital overlap among the vicinity pentacene molecules.

The output characteristics of the OTFTs with the bare PVP and the SAM on PVP are shown in Figure 5(a) and the transfer characteristics of OTFTs are shown in Figure 5(b). Detailed device properties of the fabricated OTFTs are listed in Table 2, where the field-effect

TABLE 1 The Surface Energetic Characteristics According to the SAM

| | Contact angle (water) | Contact angle (diiodo-methane) | Surface energy |
|------------|--------------------------|-----------------------------------|-------------------------|
| Bare PVP | 58.4° | 35.2° | 50.30 mJ/m ² |
| SAM on PVP | 105.3° | 58.5° | 29.93 mJ/m ² |

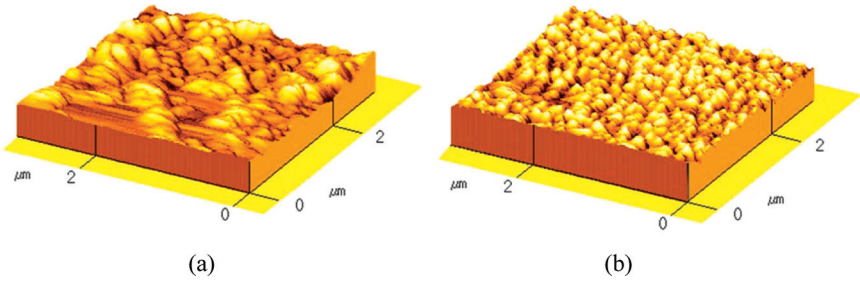


FIGURE 3 Atomic force microscopy (AFM) images of pentacene film deposited onto (a) the pristine PVP and (b) the SAM-treated PVP.

mobilities were extracted from the saturation region. From the results, it is clear that the device with the SAM-treated PVP exhibited a significant improvement in the field-effect mobility. It is likely that the observed improvement originates from the molecular ordering enhancement in the pentacene film. Consequently, it is confirmed that the ALD process can be applied to form SAM onto the gate insulator

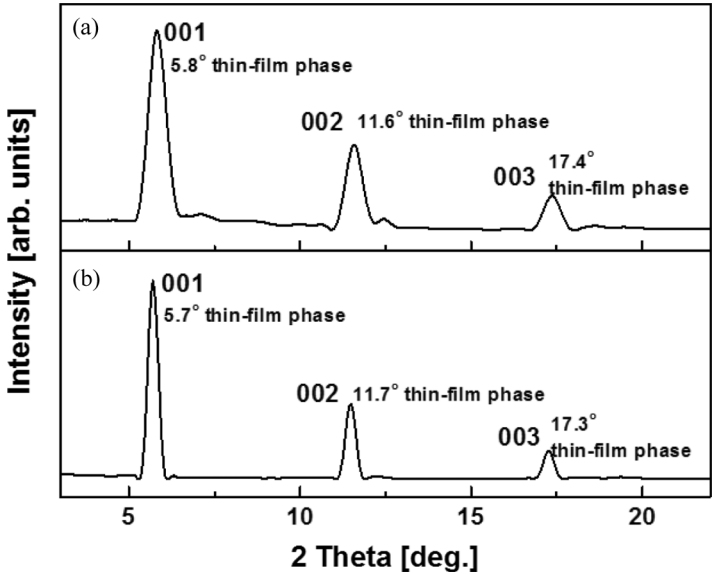


FIGURE 4 X-ray diffraction spectra of pentacene films on different substrates: (a) Pentacene/PVP/Al/Substrate and (b) Pentacene/SAM/PVP/Al/Substrate.

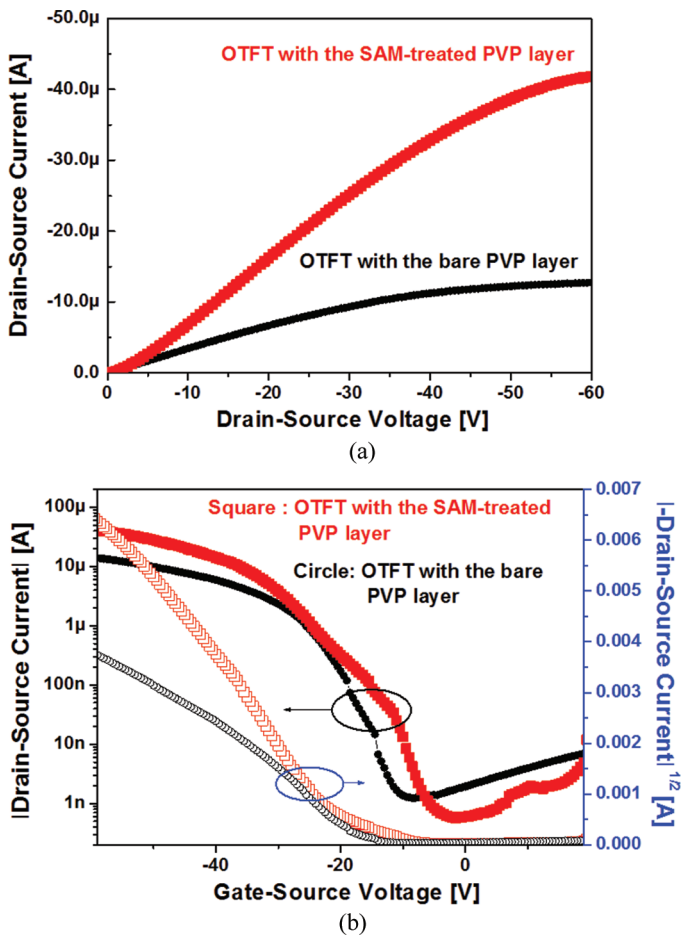


FIGURE 5 The electrical characteristics of OTFTs according to the SAM treatment: (a) the output characteristics @ $V_{GS} = -40$ V and (b) the transfer characteristics @ $V_{DS} = -40$ V.

TABLE 2 The Important Device Parameters of the Fabricated OTFTs

| | Dielectric constant of gate insulator | Threshold voltage | Subthreshold slope | On/off ratio | Field-effect mobility |
|----------------------------------|---|----------------------|-----------------------|-----------------|--------------------------------|
| OTFT with the bare PVP | ~ 3.9 | -14 V | 4.0 V/decade | $\sim 10^4$ | 0.47 cm^2/Vs |
| OTFT with the SAM-treated PVP | ~ 3.9 | -16 V | 3.1 V/decade | $\sim 10^5$ | 1.63 cm^2/Vs |

layer in OTFTs, improving the device performances by contributing to an ordered growth of pentacene molecules. Further, it is also expected that the careful control of the thickness of SAMs can be possible, thereby achieving a few-nm-thick SAMs insulator for low-voltage OTFTs with high performances.

CONCLUSION

Pentacene TFTs with the SAM-treated PVP gate insulator have been fabricated and characterized. Properties of pentacene films grown on dielectric layer are closely related with the interface conditions between the pentacene and gate dielectric layers. The hydrophilic surface of PVP layer can deteriorate the upright growth of pentacene molecules onto it. The SAM of octyltrichlorosilane deposited on the PVP in gas-phase by the ALD modifies the PVP surface more hydrophobic and affect the growth of the pentacene film, improving the morphology and crystallinity of pentacene film. Consequently, the on/off current ratio and field effect mobility could be improved.

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