

Electrical Characteristics of Organic Thin-film Transistors with Polyvinylpyrrolidone as a Gate Insulator

Jong Sun Choi*

Abstract

This paper reports the electrical characteristics of polyvinylpyrrolidone (PVPy) and the performance of organic thin-film transistors (OTFTs) with PVPy as a gate insulator. PVPy shows a dielectric constant of about 3 and contributes to the upright growth of pentacene molecules with 15.3Å interplanar spacing. OTFT with PVPy exhibited a field-effect mobility of 0.23 cm^2/Vs in the saturation regime and a threshold voltage of −12.7 V. It is notable that there was hardly any threshold voltage shift in the gate voltage sweep direction. Based on this reliable evidence, PVPy is proposed as a new gate insulator for reliable and high-performance OTFTs.

Keywords : organic thin-film transistor, polyvinylpyrrolidone, insulator

1. Introduction

There is currently much interest in organic thin-film transistors (OTFTs) due to their simple and low-temperature processability. This has expedited efforts to develop commercial applications of OTFTs in electronic devices, such as driving elements for flexible displays, radio-frequency identification tags, and large-area sensors [1-3]. The electrical characteristics of OTFTs are known to be dependent on fabrication processes, inherent properties of materials, and interfacial characteristics [4-6]. In particular, gate insulators play an important role in the performance of OTFTs and require a high dielectric constant, good interface quality, and good film morphology [7]. At an early stage of research on OTFTs, SiO_2 was the most widely used gate insulator [8] but its deposition was done via a high-temperature thermal oxidation process. Since this high-temperature process is not compatible with the mechanical flexibility and the low-cost feature of OTFTs, low-temperature and and solution-processable gate-insulating materials should be prepared for the advantages of OTFTs to be fully enjoyed. Accordingly, polymeric gate insulators such as polystyrene and poly(4-vinylphenol) are thought be a promising candidate for OTFTs by providing the simple and low-temperature process.

Polyvinylpyrrolidone (PVPy) is a unique polymer that provides remarkable properties such as good initial tack, transparency, chemical and biological inertness, very low toxicity, high media compatibility, and cross-linkable flexibility [9 and 10]. Therefore, it is believed that PVPy is suitable as a gate insulator in OTFTs. There are few reports, however, on the characteristics of PVPy and its applications to organic electronic devices. In this study, PVPy was used as a new gate insulator in OTFTs. The electrical characteristics of OTFTs with PVPy and the quality of a pentacene thin film on the PVPy layer were investigated with an atomic force microscope (AFM) and x-ray diffraction (XRD).

2. Experiments

OTFTs with a top-contact source/drain structure were fabricated. For the bottom gate electrode, an approximately 1,500Å-thick Al layer was deposited on a pre-cleaned glass substrate through the first metal shadow mask. Then, as a gate dielectric, PVPy (4 wt% in ethanol) was formed via spin-coating and baked at 100°C for 40 min in a vacuum drying oven followed by procuring at 60°C for 20 min. The PVPy layer was about 3,500 Å thick when its spinning speed and duration were optimized. After the curing process

Manuscript received November 11, 2008; accepted for publication December 12, 2008.
This work was supported by the 2006 Hongik University Research Fund.
* Member, KIDS; ** Student Member, KIDS
Corresponding Author: Jong Sun Choi
Department of Electrical, Information & Control Engineering, Hongik University 72-1 Sangsu-dong, Mapo-gu, Seoul 121-791, Korea.
E-mail: jschoi57@naver.com **Tel** : 82-2-320-3097 **Fax** : 82-2-320-1193

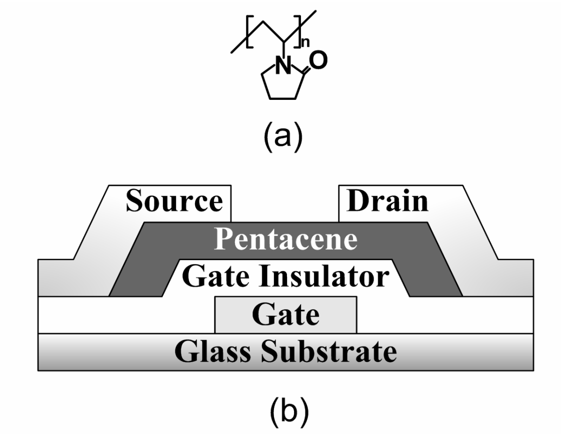

Fig. 1. (a) Molecular structure of PVPy, and (b) cross-section of the fabricated top-contact OTFT.

was completed, a 600Å-thick pentacene layer, as an organic semiconductor, was thermally evaporated through the second shadow mask. Pentacene (Tokyo Kasei Kogyo Co., Ltd.) was used without further purification and deposited at a rate of 1.0 Å/s. A 400Å-thick source and drain electrodes on top of the pentacene were thermally evaporated through the third shadow mask, with a channel length (L) of 90 µm and a width (W) of 300 µm. The evaporation process was carried out under a base pressure of about 1.6×10^{-6} Torr. Fig. 1 shows the molecular structure of PVPy and a schematic diagram of the fabricated OTFT.

The crystallinity of each layer was studied via XRD (DMAX 2500, Rigaku) with a monochromatic Cu Kα (λ = 1.54 Å), and the surface morphology of the pentacene film was examined under an AFM (XE-150, PSIA, Inc.), using the contact mode. The dielectric property of PVPy and the electrical characteristics of OTFTs were measured with an impedance analyzer (HP 4192LF, Agilent Technologies) and a semiconductor analyzer (EL 421C, Elecs Co.), respectively.

3. Results and Discussion

The device for the capacitance measurement consisted of a PVPy layer sandwiched between Al and Au electrodes, and the dielectric constant was calculated using Eq. 1 [11] below:

$$C = \frac{\varepsilon_0 \varepsilon_r}{d} A, \qquad \text{Eq. 1}$$

wherein C is the measured capacitance, ε_0 is the permittivity of free space, ε_r is the relative dielectric constant of the insulator, A is the area of the capacitor, and d is the insulator thickness. Fig. 2 (a) shows the dielectric constants as functions of the applied frequency. The dielectric constant of PVPy was about 3 at 100 kHz, which is in the range of conventional polymeric insulators, such as polystyrene and poly(vinyl acetate). The insulation property of the PVPy gate insulator was also examined using the Al electrode/PVPy (3,500 Å)/Al electrode structure, as shown in Fig. 2 (b). The fabricated PVPy film exhibited an electric field strength of about 2 MV/cm.

The structural and morphological characteristics of the pentacene film deposited onto the PVPy layer are shown in Fig. 3. The XRD spectrum showed two diffraction peaks at 5.76° and 6.14°, which corresponded to the thin-film phase and the triclinic bulk phase, respectively. The strongest peak near 5.76° indicated that the major component of this film was a thin-film phase with an interplanar spacing of

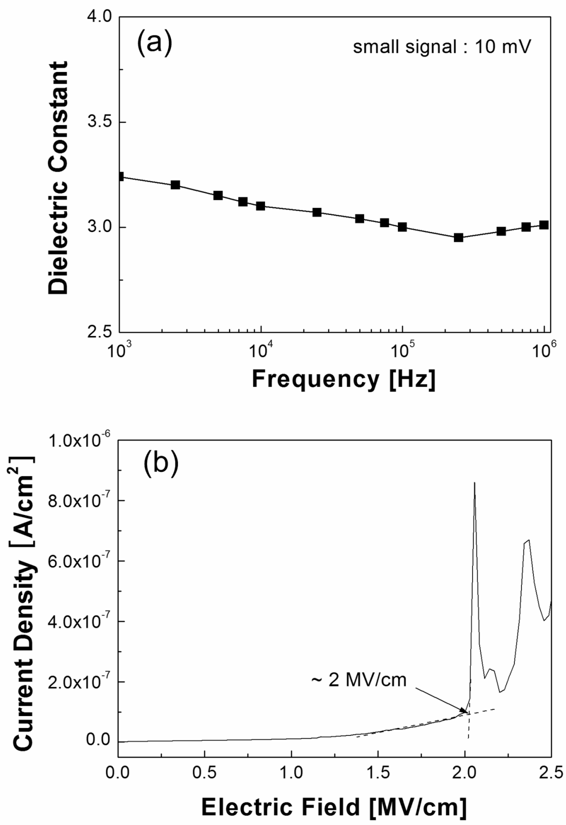

Fig. 2. (a) Calculated dielectric constants of PVPy at various frequencies, and (b) current leakage of the 3,500Å-thick PVPy insulator as a function of the electric field.

15.3 Å, whereas the diffraction peak near 6.14° corresponded to the bulk phase with an interplanar spacing of 14.4 Å. Considering that the pentacene molecules were about 16.5 Å long, it was found that they were highly ordered in the direction perpendicular to the PVPy layer. The AFM image obviously shows that pentacene molecules tend to form well-ordered crystallites in a herringbone structure, as shown in the inset of Fig. 3, where the grain size was 600-800 nm. Since the interaction of π-electron systems between adjacent molecules depends strongly on their stacking nature, it is expected that the vertical alignment of the pentacene molecules to the PVPy-coated substrate can provide a strong π-orbital overlap and increase the charge transport properties.

Fig. 4 (a) shows the drain current (I_D) versus the drain voltage (V_D) curves of the OTFT with the PVPy gate insulator with different negative gate voltages (V_G). It was observed that the device exhibited a good saturation behavior and that gate voltages clearly modulated its current enhancement. The corresponding plot of $|I_D|^{1/2}$ versus V_G that was obtained at the V_D of -30 V is shown in Fig. 4 (b). The field-effect mobility (μ_{eff}) was calculated in the saturation region using Eq. 2 [12], as follows:

$$I_D = \frac{W \mu_{eff} C_i}{2L} (V_G - V_T)^2, \qquad \text{Eq. 2}$$

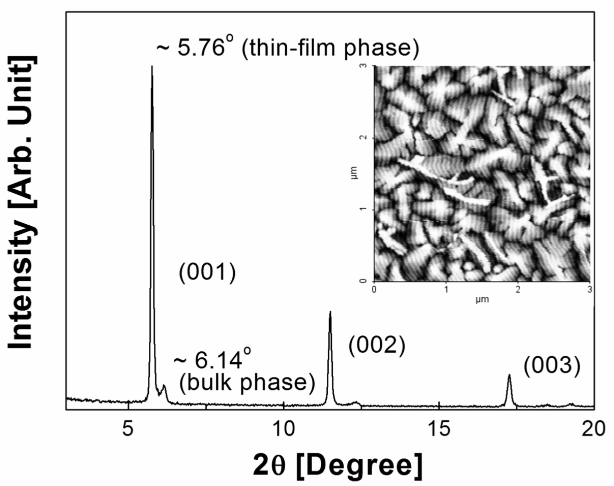

Fig. 3. (a) XRD pattern of the 1,200Å-thick pentacene film on the PVPy gate insulator. The inset shows the surface of the 300Å-thick pentacene film deposited on the PVPy gate insulator characterized by AFM (3 μm x 3 μm).

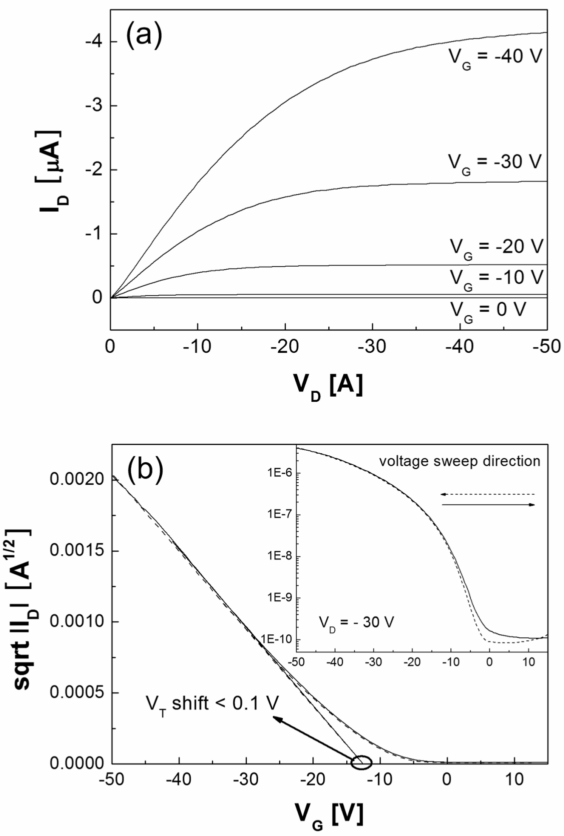

Fig. 4. Electrical characteristics of the fabricated OTFT with the PVPy gate insulator: (a) output characteristics with varying V_G, and (b) transfer curve at V_D = -30 V. The inset in (b) shows the $\mathrm{Log}_{10}|I_D|$ versus V_G plot.

wherein C_i is the capacitance of the gate insulator per unit area, and V_T is the threshold voltage. The calculated field-effect mobility and the extracted threshold voltage were 0.23 cm^2/Vs and -12.7 V, respectively. The on/off current ratio was about 5×10^4, with a subthreshold slope of 3.2 V/decade, as shown in the inset of Fig. 4 (b). It is thought that the presented on/off current ratio is relatively low due to the gate leakage current through the PVPy insulator layer, which, it is believed, can be modified by optimizing the thickness of the PVPy layer. Although the TFT with the PVPy gate insulator could not establish significant progress, the reported device performance values are decent compared with those of devices with polymeric gate insulators. Of particular interest is the stable operation without a shift in the threshold voltage in the gate voltage sweep direction [see Fig. 4 (b)], because OTFTs that adopt polymeric gate insulators often suffer from a threshold voltage shift. A recent study demonstrated that the threshold voltage shift is closely related to the charge trapping phenomenon at the interface between organic semiconductor and gate insulator layers [13]. Based on the said study, it was inferred that the PVPy gate insulator formed a defect-free interface and thus contributed to the stable operation with a threshold voltage shift of less than 0.1 V. This property can make PVPy a unique candidate for polymeric gate insulation to achieve reliable OTFTs on flexible substrates.

4. Conclusion

In this study, PVPy was proposed as a new gate insulator for OTFTs. The experimental results showed that PVPy has a dielectric constant of about 3 and that the pentacene film deposited on the PVPy layer formed a strong thin-film phase with an interplanar spacing of 15.3 Å. The OTFT with the PVPy gate insulator exhibited a field-effect mobility of 0.23 cm/Vs, a threshold voltage of -12.7 V, and an on/off current ratio of $5x10^4$ with a subthreshold slope of 3.2 V/decade. In particular, the device showed a stable operating performance without shifts in the threshold voltage in the gate voltage sweep direction. Based on the author's first-hand knowledge, this is the first report that studied the characteristics of OTFTs with the PVPy gate insulator. In addition to the presented peculiarities of PVPy--that is, its transparency and cross-linkable flexibility--its availability as a gate insulator for high-performance OTFTs was also shown. It is believed that this study can help researchers in the field of organic electronics. Further studies are in progress to improve the device performance and develop a cross-linking process below 150°C.

References

[1] S.R. Forrest, *Nature*, **428**, 911 (2004).
[2] R. Rotzoll, S. Mohapatra, V. Olariu, R. Wenz, M. Grigas, K. Dimmler, O. Scchekin, and A. Dodabalapur. *Appl. Phys. Lett.*, **88**, 123 502 (2006).
[3] L. Wang, D. Fine, S. I. Khondaker, T. Jung, and A. Dodabalapur. *Sens. Actuators*, **B113**, 539 (2006).
[4] D. R. Hines, V. W. Ballarotto, E. D. Williams, Y. Shao, and S. A. Solin. *J. of Appl. Phys.*, **101**, 024 503 (2007).
[5] A. Facchetti, M. H. Yoon, and T. J. Marks. *Adv. Mater.*, **17**, 1705 (2005).
[6] K. P. Pemstich, S. Haas, D. Oberhoff, C. Goldmann, D. J. Gundlach, B. Batlogg, A. N. Rashid, and G. Schitter. *J. of Appl. Phys.*, **96**, 6431 (2004).
[7] J. H. Sung, S. J. Park, J. H. Park, H. J. Choi, and J. S. Choi. *Synth. Met.*, **156**, 861 (2006).
[8] Z. Bao, A. Dodabalapur, and A. J. Lovinger. *Appl. Phys. Lett.*, **69**, 4108 (1996).
[9] W. Xu, H. Chen, H. Li, and M. Wang. *Colloids Surf. A*, **266**, 68 (2005).
[10] J. Swei and J.B. Talbot, *J. Appl. Polym. Sci.*, **90**, 1156 (2003).
[11] R. F. Pierret. *Field Effect Devices* (Addison-Wesley, New Jersey, 1983), p.45.
[12] S. M. Sze and K. K. Ng. *Physics of Semiconductor Devices* (John Wiley & Sons, New Jersey, 2007), p. 306.
[13] Jiyoul Lee, J. H. Kim, Seongil Im, and D. Y. Jung. *J. of Appl. Phys.*, **96**, 2301 (2004).