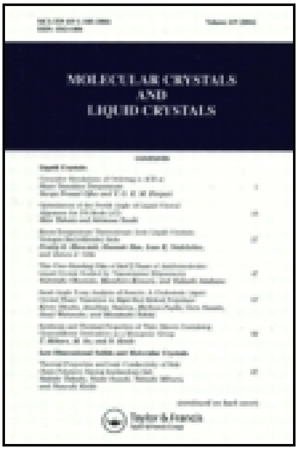

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

The Photo Cross-Linking Effect of a Polymeric Insulator on an Organic Thin-Film Transistor

Won-Ho Kim [a], Jin-Hyuk Bae [a] & Sin-Doo Lee [a]

[a] School of Electrical Engineering, Seoul National University, Kwanak, Seoul, Korea

To cite this article: Won-Ho Kim, Jin-Hyuk Bae & Sin-Doo Lee (2007): The Photo Cross-Linking Effect of a Polymeric Insulator on an Organic Thin-Film Transistor, Molecular Crystals and Liquid Crystals, 471:1, 155-162

To link to this article: http://dx.doi.org/10.1080/15421400701548092

Mol. Cryst. Liq. Cryst., Vol. 471, pp. 155–162, 2007
Copyright © Taylor & Francis Group, LLC
ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400701548092

The Photo Cross-Linking Effect of a Polymeric Insulator on an Organic Thin-Film Transistor

Won-Ho Kim
Jin-Hyuk Bae
Sin-Doo Lee
School of Electrical Engineering, Seoul National University, Kwanak, Seoul, Korea

We studied how the photo cross-linking density of a polymeric insulator affects the mobility of organic thin-film transistors (OTFTs). The photo cross-linking density was monitored in terms of the optical anisotropy with changing the exposure time of the linearly polarized ultraviolet (LPUV) light. It was found that the increase in the mobility with the LPUV exposure time results mainly from the high packing density of pentacene molecules through the photo cross-linking of the polymer. This suggests that the packing density of pentacene molecules plays a more essential role in the enhancement of the mobility than the alignment of pentacene molecules.

Keywords: optical anisotropy; organic thin-film transistors; photo cross-linking density

INTRODUCTION

Organic thin-film transistors (OTFTs) have been extensively investigated for potential applications in flexible displays, various sensors, and radio frequency identification cards [1–3]. For the OTFTs, the polymer materials are promising for gate insulators since they have many advantages such as low cost, large area fabrication, and low temperature processing. For example, poly (vinyl cinnamate) (PVCi) has often been used as a polymeric insulator on the OTFT due to its low surface roughness, low leakage current, and small hysteresis [4]. In addition to above advantages, the PVCi has an interesting property

---

This work was supported in part by the Ministry of Science and Technology of Korea through the 21st Century Frontier Research and Samsung Electronics, AMLCD.

Address correspondence to Prof. Sin-Doo Lee, School of Electrical Engineering, Seoul National University, Kwanak P.O. Box 34, Seoul 151-600, Korea. E-mail: sidlee@plaza.snu.ac.kr

of photo-alignment of liquid crystal (LC) molecules [5]. The PVCi induces an anisotropic surface energy by cross-linking the excited side chains upon the linearly polarized ultraviolet (LPUV) exposure [6]. Although the anisotropic surface energy of the PVCi layer strongly affects the interfacial forces between the PVCi molecules and other molecules deposited on the PVCi layer, most researches have never been explored the photo cross-linking effect of the PVCi molecules on the electrical performances of the OTFTs. Thus, the underlying mechanism for the enhancement of the mobility associated with the photo cross-linking process has not been fully understood so far.

In this work, we study how the photo cross-linking density of the PVCi layer affects the mobility of the pentacene-based OTFT. The photo cross-linking process was carried out by the LPUV exposure onto the PVCi layer. The cross-linking density was monitored in terms of the magnitude of the optical anisotropy with changing the exposure time of the LPUV. The mobility of the OTFT with the PVCi insulator layer treated by the LPUV was found to be proportional to the photo cross-linking density. It should be emphasized that no alignment of pentacene molecules was observed on the photo cross-linked PVCi layer by the LPUV exposure.

EXPERIMENTAL

A top-contact OTFT structure fabricated in our study is shown in Figure 1. Indium-tin-oxide (ITO) on a glass substrate, etched by photo-lithography, was used as a gate electrode. The glass substrate with the ITO pattern was cleaned with acetone, isoprophyl alcohol,

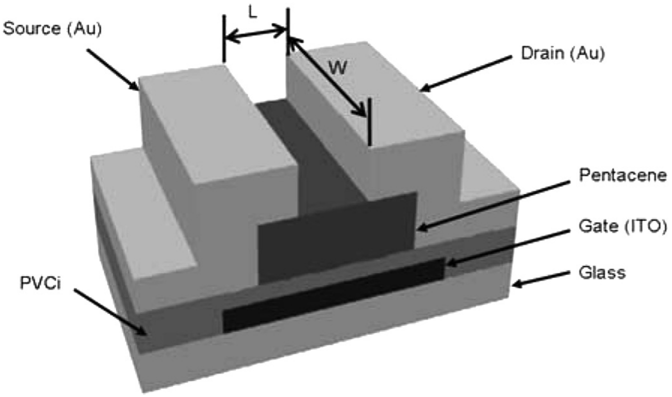

FIGURE 1 The basic structure of a top-contact OTFT.

methanol, and de-ionized water in sequence to remove residual impurities in a ultrasonicator. For preparing a gate dielectric insulator layer, the PVCi was dissolved in cyclopentanone in 10 wt.%. It was spin coated onto the ITO glass substrate and cured at 60°C for 5 hours. The thickness of the PVCi layer was 570 nm. In order to induce the photo cross-linking, the LPUV was exposed on the PVCi layer as a function of time. The UV source was a high pressure Hg lamp with the intensity of 6.3 mW/cm^2. The cross-linking density of the PVCi layer was monitored in terms of the magnitude of the optical anisotropy using a photo-elastic modulation (PEM) technique [7]. The pentacene material used as an active layer was purchased from TCI Ltd., and it was thermally deposited through a shadow mask onto the PVCi layer at the rate of 0.5 Å/sec. The thickness of the pentacene layer was 60 nm. The source electrode and the drain electrode were subsequently prepared on the pentacene layer using Au. All the deposition processes were carried out under the base pressure of approximately 10^{-6} Torr. During the deposition, the substrate was fixed at room temperature. The thickness of the Au electrode was 80 nm. The length and the width of the channel were 50 μm and 1 mm, respectively. The electrical properties of the pentacene-based OTFT were measured using the 4155 A semiconductor parameter analyzer under an ambient pressure at room temperature.

RESULTS AND DISCUSSION

The photo cross-linking process of the PVCi molecules upon the LPUV exposure was shown in Figure 2. The LPUV exposed onto the PVCi

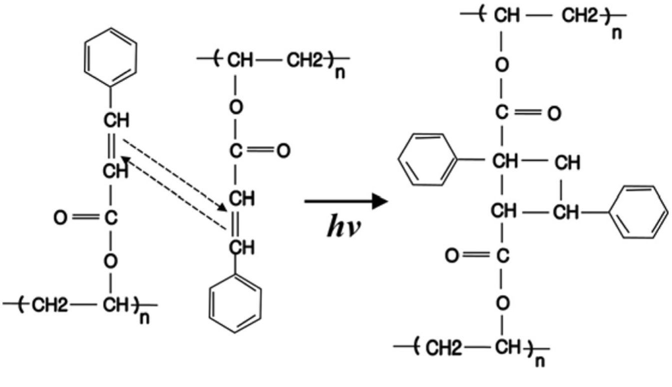

FIGURE 2 The molecular structure of the PVCi and a dimerized PVCi by photo cross-linking upon the LPUV exposure.

layer breaks the C=C bonds of cinnamate side chains that are parallel to the direction of the LPUV. The broken side chains adjacent to main chains cause [2 + 2] cycloaddition, and then create an anisotropic distribution of cross-linked molecules along the direction perpendicular to the LPUV [8].

The refractive index of the polymer material can be empirically given as the Gladstone-Dale expression [9].

$$n - 1 = \frac{d}{M} R_m, \tag{1}$$

where n is the refractive index, M is the molar mass of the monomer unit, d is the molecular density, and R_m is the monomer molar refractive index. Note that the molecular density of molecules (d) varies with the refractive index because the photo-reaction disturbs only the distribution of the PVCi molecules. While the LPUV depletes the molecules in the direction parallel to the LPUV, the density of polymer molecules along the direction perpendicular to the LPUV increases with the photo cross-linking as shown in Figure 2. Consequently, the induced optical anisotropy of the PVCi layer is determined by the difference in the molecular density between the parallel orientation and the perpendicular orientation to the LPUV direction.

The PEM experimental set up to measure the optical anisotropy is shown in Figure 3. The optical anisotropy Δ_n of the PVCi layer, the difference between the ordinary refractive index n_o and the extraordinary refractive index n_e, results in the phase retardation $\Delta\phi$ as [10]

$$\Delta\phi = 2\pi \frac{t}{\lambda} (n_o - n_e), \tag{2}$$

where t is the thickness of the PVCi layer and λ is the wavelength of the probe light. From the measured phase retardation $\Delta\phi$ as a function of θ for the rotation angle, the optical anisotropy Δn was determined.

Figure 4 shows the measured phase retardations of both the PVCi layer and the pentacene layer for several different LPUV exposure times together with the corresponding output characteristic curves of our OTFTs. As shown in Figure 4(a), no anisotropy in the molecular distribution was observed in the PVCi layer without the LPUV exposure. Moreover, the optical anisotropy of the pentacene layer itself, deduced from the data for both the PVCi layer and the pentacene layer, is negligible. It means that there is no substantial alignment of pentacene molecules along any direction. The OTFT with such isotropic PVCi insulator shows the lowest drain current, which is about − 3.6 μA at the drain voltage of −60 V and the gate voltage of − 60 V. The phase retardations of the PVCi insulator and the

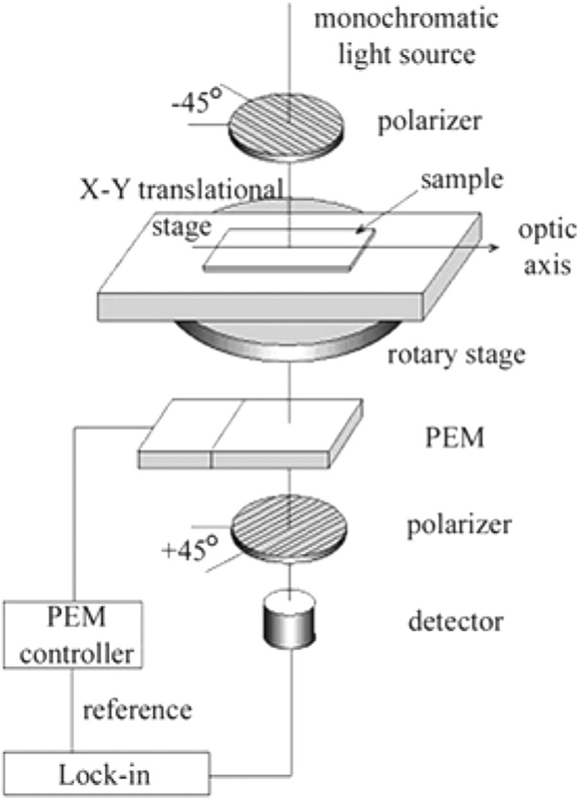

FIGURE 3 The PEM experimental setup to measure the optical anisotropy of the PVCi layer and the pentacene layer.

pentacene layer together with the output characteristic curve for the LPUV exposure time of 600 sec are shown in Figure 4(b). It is clear that the optical anisotropy of the pentacene layer itself is negligible although the phase retardation of the PVCi layer is about 0.01 rad. In contrast to the LC molecules, no alignment of the pentacene molecules is induced on the LPUV exposed PVCi layer. This implies that the LPUV exposure on the PVCi insulator influences only the packing density of the pentacene molecules. **The induced optical anisotropy of the PVCi upon the exposure of the LPUV increases with increasing cross-linking density of the PVCi [9]. As a result, the interaction sites at the interface between the PVCi layer and the pentacene layer are increased. It brings the higher packing density of pentacene molecules**. The packing

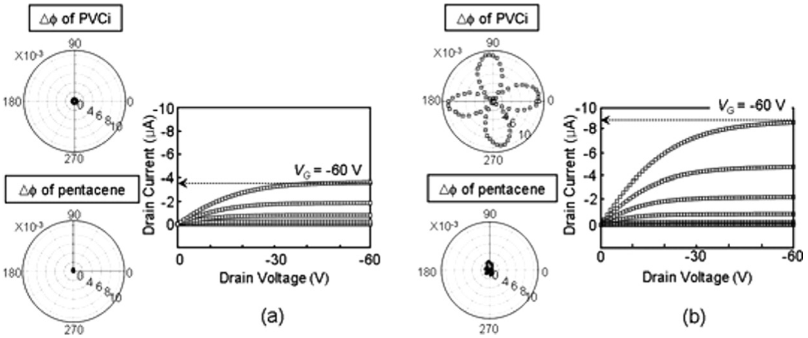

FIGURE 4 The phase retardations of both the PVCi layer and the pentacene layer, and the output characteristic curves of our OTFTs: (a) 0 sec, (b) 600 sec for the LPUV exposure time. The gate voltages are varied in a step of $-10\,\text{V}$ from $0\,\text{V}$ to $-60\,\text{V}$.

density of the pentacene molecules is predominantly governed by the cross-linking density of the PVCi molecules which is proportional to the optical anisotropy as expected from Eq. (1).

We now calculate the field effect mobility in the saturation regime ($V_D = -60\,\text{V}$) from the following equation [11].

$$I_{D,sat} = \frac{W}{2L} C_i \mu (V_G - V_T)^2, \tag{3}$$

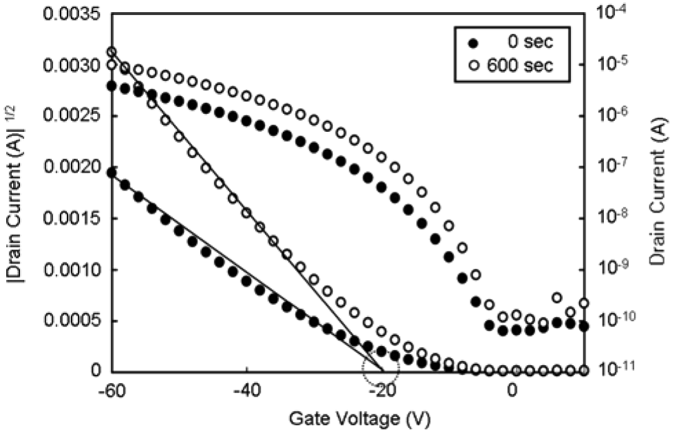

FIGURE 5 The transfer characteristic curves of the OTFTs: filled circles and open circles represent the LPUV exposure times of 0 sec and 600 sec, respectively. The drain-source voltage of $-60\,\text{V}$ was used to calculate the mobility.

where C_i is the capacitance of insulator per unit area, L is the length of the channel, W is the width of the channel, and V_T is the threshold voltage. From the characteristic curves in Figure 5, the mobilities for the LPUV exposure times of 0 sec and 600 sec were determined as $0.052\,\mathrm{cm}^2/\mathrm{Vs}$ and $0.111\,\mathrm{cm}^2/\mathrm{Vs}$, respectively, using C_i of $5.9\,\mathrm{nF/cm}^2$. Note that the threshold voltage remains almost same as $-20\,\mathrm{V}$ and no substantial difference in the current on-off ratios 10^5 was observed. **Moreover, the mobilities in two OTFTs, in one of which the carrier flow direction is parallel to the direction of the LPUV and the other perpendicular to the LPUV direction, show no substantial difference ($<$15%) [12].** It is concluded that the mobility is proportional to the optical anisotropy of the PVCi layer through the packing density of the pentacene molecules, not through the alignment of the pentacene molecules. This is evident from the fact that no optical anisotropy in the pentacene layer itself was observed for the LPUV exposure time of 600 sec.

CONCLUSION

We observed that the photo cross-linked molecules of the PVCi insulator strongly affects the mobility of the OTFT through the packing density of the pentacene molecules. Although the optical anisotropy in the PVCi insulator was induced upon the LPUV exposure, no optical anisotropy in the pentacene layer itself was observed. In contrast to the LC molecules, the pentacene molecules are influenced by the LPUV exposure through the packing density, not through the alignment. The photo cross-linking density of the PVCi insulator plays an important role in the magnitude of the mobility of the OTFT by increasing the packing density of the pentacene molecules.

REFERENCES

[1] Dimitrakopoulos, C. D. & Malenfant, P. R. L. (2002). *Adv. Mater.*, *14*, 99.

[2] Horowitz, G. (1998). *Adv. Mater.*, *10*, 365.

[3] Zhu, Z.-T., Mason, J. T., Dieckmann, R., & Malliaras, G. G. (2002). *Appl. Phys. Lett.*, *81*, 4643.

[4] Stadlober, B., Zirkl, M., Beutl, M., & Leising, G. (2005). *Appl. Phys. Lett.*, *86*, 242902.

[5] Schadt, M., Schmitt, K., Kozinkov, V., & Chigrinov, V. (1992). *Jpn. J. Appl. Phys.*, *31*, 2155.

[6] Ionescu, A. Th., Barberi, R., Giocondo, M., Iovane, M., & Alexe-Ionescu, A. L. (1998). *Phys. Rev.* E, *58*, 1967.

[7] Lee, J.-H., Yu, C.-J., & Lee, S.-D. (1998). *Mol. Cryst. Liq. Crys.*, *321*, 317.
[8] Egerton, P. L., Pitts, E., & Reiser, A. (1981). *Macromolecules*, *14*, 95.
[9] Assaid, I., Bosc, D., & Hardy, I. (2004). *J. Phys. Chem. B.*, *108*, 2801.
[10] Yariv, A. & Yeh, P. (1984). *Optical Wave in Crystals*, John Wiley & Sons: New York.
[11] Sze, S. M. (1981). *Physics of Semiconductor Devices*, Wiley: New York.
[12] Bae, J.-H., Kim, J., Kim, W.-H., & Lee, S.-D. (2007). *Jpn. J. Appl. Phys.*, (in press).