

An Organic Electrophosphorescent Device Driven by All-Organic Thin-Film Transistor using Polymeric Gate Insulator

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Abstract

In this paper, we demonstrate that the organic electrophosphorescent device is driven by the organic thin film transistor with spin-coated photoacryl gate insulator. It was found that electrical output characteristics in our organic thin film transistors using the staggered-inverted top-contact structure showed the non-saturated slope in the saturation region and the sub-threshold nonlinearity in the triode region, where we obtained the maximum power luminance that was about 90 cd/m². Field effect mobility, threshold voltage, and on-off current ratio in 0.45 μm thick gate dielectric layer were 0.17 cm²/Vs, -7 V, and 10⁶, respectively. In order to form polyimide as a gate insulator, vapor deposition polymerization process was also introduced instead of spin-coating process, where polyimide film was co-deposited by high-vacuum thermal evaporation from 4,4'-oxydiphthalic anhydride (ODPA) and 4,4'-oxydianiline (ODA) and cured at 150°C for 1hr. It was also found that field effect mobility, threshold voltage, on-off current ratio, and sub-threshold slope with 0.45 μm thick gate dielectric films were 0.134 cm²/Vs, -7 V, and 10⁶ A/A, and 1 V/decade, respectively.

Keywords : Organic electrophosphorescent device, organic thin film transistors, vapor deposition polymerization process

1. Introduction

Organic electroluminescent devices (OLEDs) have been investigated for many years in search for a possible use in full color panel display applications and backlights, and organic materials have drawn attention in the application of conductor and semiconductor in recent years. It was not until recently, however, that these devices could be fabricated by organic thin film

transistors (OTFTs) as an active matrix device for operating to large area display. The use of active matrix organic electroluminescent displays allows polymeric and plastic substrates to replace the glass substrates currently. Polymeric and plastic substrates have many advantages in terms of weight and ruggedness over conventional glass substrates [1,2]. In this paper, we demonstrated electrophosphorescent devices with all-organic thin-film transistors using organic material as a gate insulator. Also, we propose a new processing method of polymeric gate dielectric to be applied in thin-film field-effect transistors. Vapor deposition polymerization, which is one of the most useful methods for producing high performance polymer thin film, is introduced to form the dielectric. This method is appropriate for mass production in various end-user applications, for example, flat panel display, because it has the advantage of shadow mask patterning and in-situ dry processing with flexible and large-area substrate.

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2. Experiments

1. Organic thin-film transistor through spin-coating process and organic electrophosphorescent device

In this study, we demonstrate electrophosphorescent devices with all-organic thin-film transistors using organic material as a gate insulator. We studied various organic thin film transistor electrophosphorescent devices using phosphorous materials that have achieved high brightness and good quantum efficiency. Here, we describe high efficiency OLEDs employing the green electrophosphorescent material fac tris(2-henylpyridine) iridium [Ir(ppy)₃], doped into various electron transporting layer hosts.

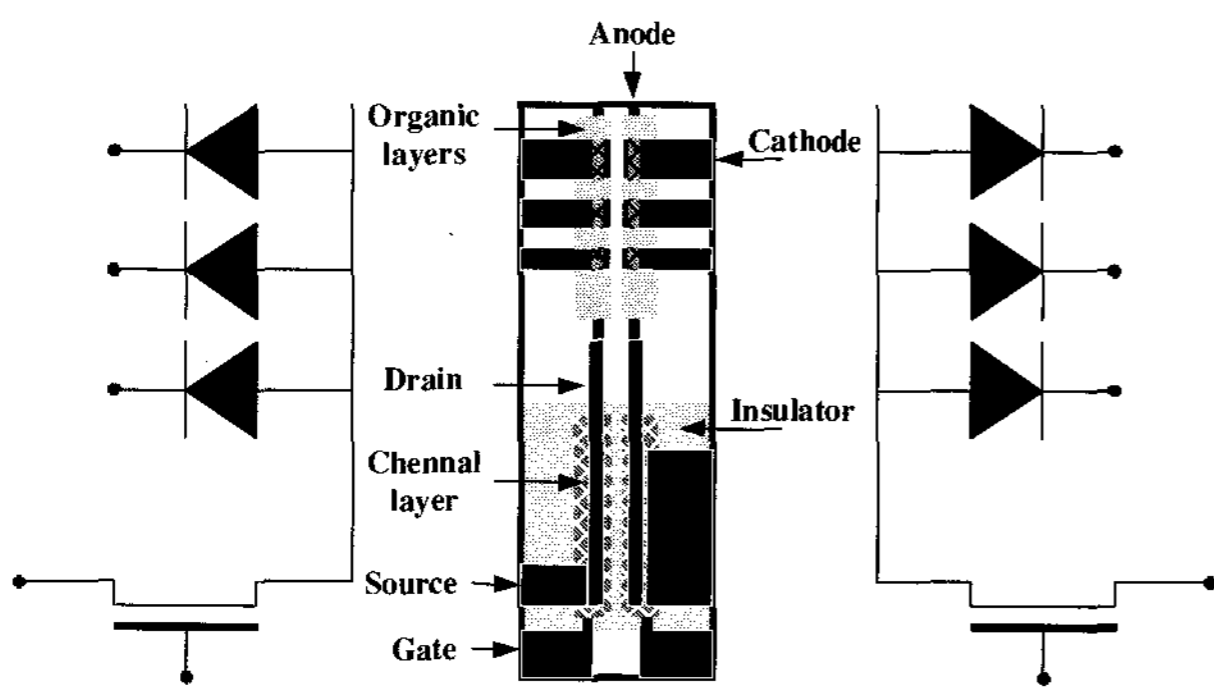


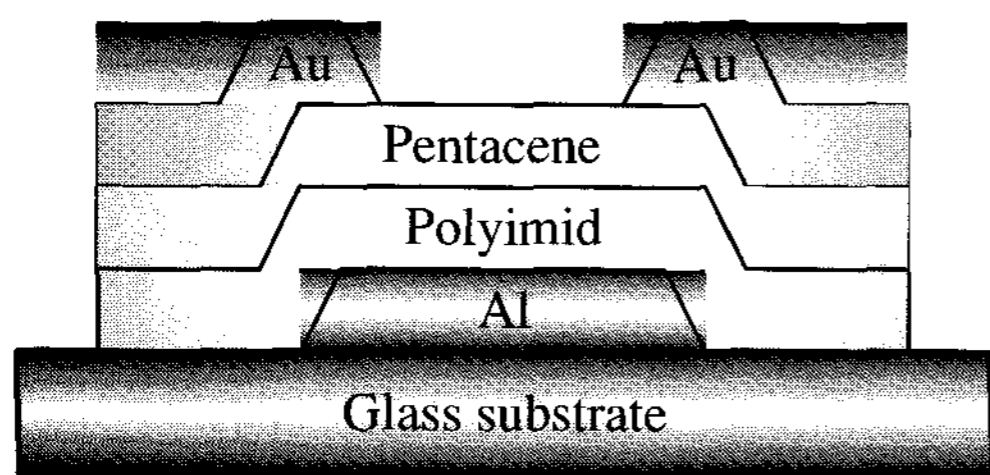
Fig. 1. Schematic of the cross section of the organic electrophosphorescent device derived organic thin film transistor using photoacryl as a gate insulator.

First, gold was deposited as a drain electrode of TFT and an anode of OLEDs. However, the device failed to function properly because of injecting much more holes into anode using Au(gold), which has lower work function than indium tin oxide. Hence patterned ITO was used as an anode for connecting with drain. The devices were completed by thermal deposition of Au to form the source and drain contacts through a shadow mask. Second, photoacryl (thickness $\sim 0.5 \mu\text{m}$) was used as a gate insulator, gold (Au) and aluminum (Al) were used as contact electrodes. And pentacene (thickness $\sim 80 \text{ nm}$) was deposited as channel layers of OTFTs with W/L ratio 156. Since pentacene is a small molecule and is insoluble, it can be thermally deposited in vacuum. Au, as the source and drain electrodes, has a high work function, which has been proven to give a good ohmic

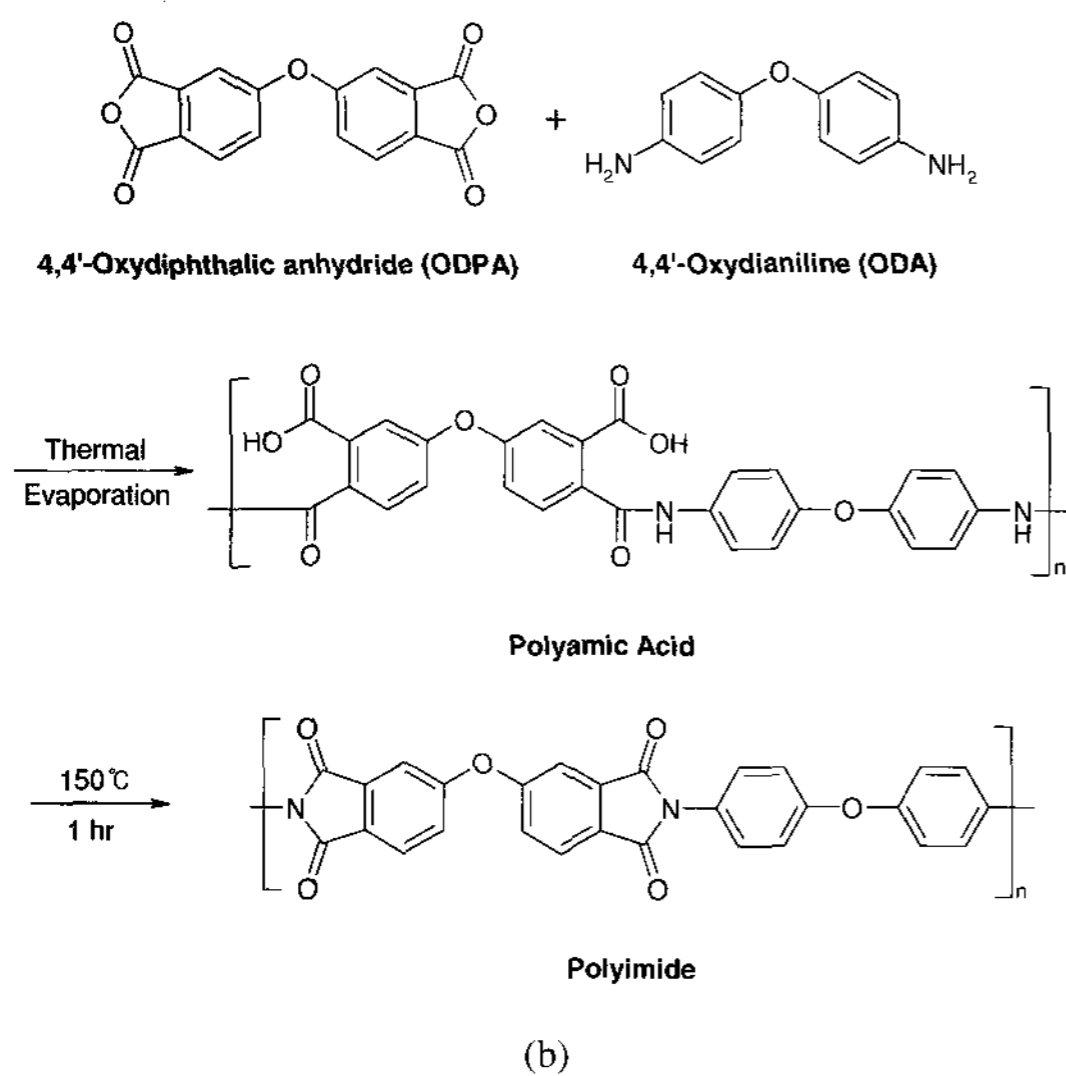
contact to pentacene [3]. The staggered-inverted top-contact structure, Al/photoacryl/pentacene/Au, was fabricated. Fig. 1 shows the device scheme of electrophosphorescent devices with all-organic thin-film transistor. Photoacryl solutions were filtered through a $0.45\text{-}\mu\text{m}$ PURADISC 25 PP FILTER and spun onto the glass at $2000\sim 6000 \text{ rpm}$ for 30 sec in an inert atmosphere. Photoacryl film of about $\sim 1.5 \mu\text{m}$ -thick was spin-coated and cured at 220°C for 1 hour. Its breakdown field measured to be larger than 1 MV/cm . Pentacene active layer was deposited by thermal evaporation at $5\times 10^{-7} \text{ Torr}$, deposition rate of $0.2 \sim 0.3 \text{ \AA/s}$, and total thickness of 80 nm after and going a material purification process by vacuum gradient sublimation. Pentacene, a short-chain molecule consisting of five linearly fused benzene rings, is typically used as a *p*-type semiconductor. Pentacene was obtained from commercial sources and purified by vacuum temperature-gradient sublimation prior to deposition [7,8]. Improved purity of the semiconductor material, room temperature, and low deposition rates has been used to improve the performance of organic thin-film transistor. Finally, 8-hydroxyquinolino (Alq₃) was deposited as electron transporting material, triphenyl diamine TPD as hole transporting material, 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) as host material, Ir(ppy)₃ as dopant material (8%), lithium aluminum (Li:Al) as a cathode, and indium-tin-oxide (ITO) as an anode for OLEDs, respectively. The structure of electrophosphorescent diode was ITO/TPD/BCP:Ir(ppy)₃/BCP/Alq₃/Li-Al/Al.

2. Organic thin-film transistor through vapor deposition polymerization process.

In the new process, 140-nm -thick indium-tin-oxide electrode as a gate insulator was patterned on the glass substrate by photolithography. Following the polyimide formation, a 178-nm -thick aluminum electrode was thermally deposited onto the polyimide film. Its breakdown field was larger than 0.32 MV/cm . All-organic thin-film transistors were fabricated to demonstrate that thermal evaporated polyimide was used as a gate dielectric. All transistors fabricated on glass substrates had a staggered-inverted structure as shown in Fig. 2-(a). In this structure, two different voltage sources were used; one is cross the dielectric layer (V_G)



(a)



(b)

Fig. 2. Schematic of transistor using VDP as a gate insulator(a), and polyimides by the presence of the imide functionality, a cyclic tertiary amine bound by two carbonyl groups, and either an aliphatic or aromatic group in the main chain(b).

generating the charges (charge injection into the channel), and the other is along the active layer (channel) (V_D) to drive them from source to drain.

Polyimide is a polymer made from the condensation polymerization of an acid dianhydride and a diamine. It is characterized by the presence of the imide functionality, a cyclic tertiary amine bound by two carbonyl groups (Fig. 2-(b)), and either an aliphatic or aromatic group in the main chain. Typically, the synthesis of polyimide consists of two stages. In the first stage, the precursor of the imide moiety, a polyamic acid, is formed by a polycondensation reaction of an acid dianhydride with a difunctional base (a primary amine). During the second stage, the polyamic acid is converted to polyimide when sufficient heat is supplied to remove the solvent and to initiate ring closure with intramolecular expulsion of water. The curing process and the associated cross-linking affect the properties of polyimide mechanically, thermally, and electrically. This heat treatment, known as

“curing,” usually has a final temperature range of 150 ~ 300 °C. The two-stage synthesis described here has been widely used as it brings about isolation of a polyamic acid, the soluble product from the first-stage. Currently, the bulk of the material received by end users for polyimide application in IC processing is in the polyamic acid form, which undergoes conversion to polyimide only after the wafer is applied. We doped ODA (4,4'-oxydianiline) and ODPA (4,4'-oxydiphthalic anhydride) by high vacuum (5×10^{-7} Torr) thermal evaporation. The doping concentration was about 50 % ODA from the experiment. Then, the device was cured at 150 °C for 1 hour and showed FT-IR graph of the polyimide. Aluminum was used for the gate metal, and the pentacene active layer was deposited by thermal evaporation at 5×10^{-7} Torr, deposition rate of 0.2 ~ 0.3 Å/s, and total thickness of 80 nm after the material purification by vacuum gradient sublimation. During the deposition of pentacene, the substrates were held at room temperature. Improved purity of the semiconductor material, moderate heating, and low deposition rates were maintained to improve the performance of thin-film transistor [4,6]. The devices were completed by thermal deposition of Au to form the source and drain contacts through a shadows mask.

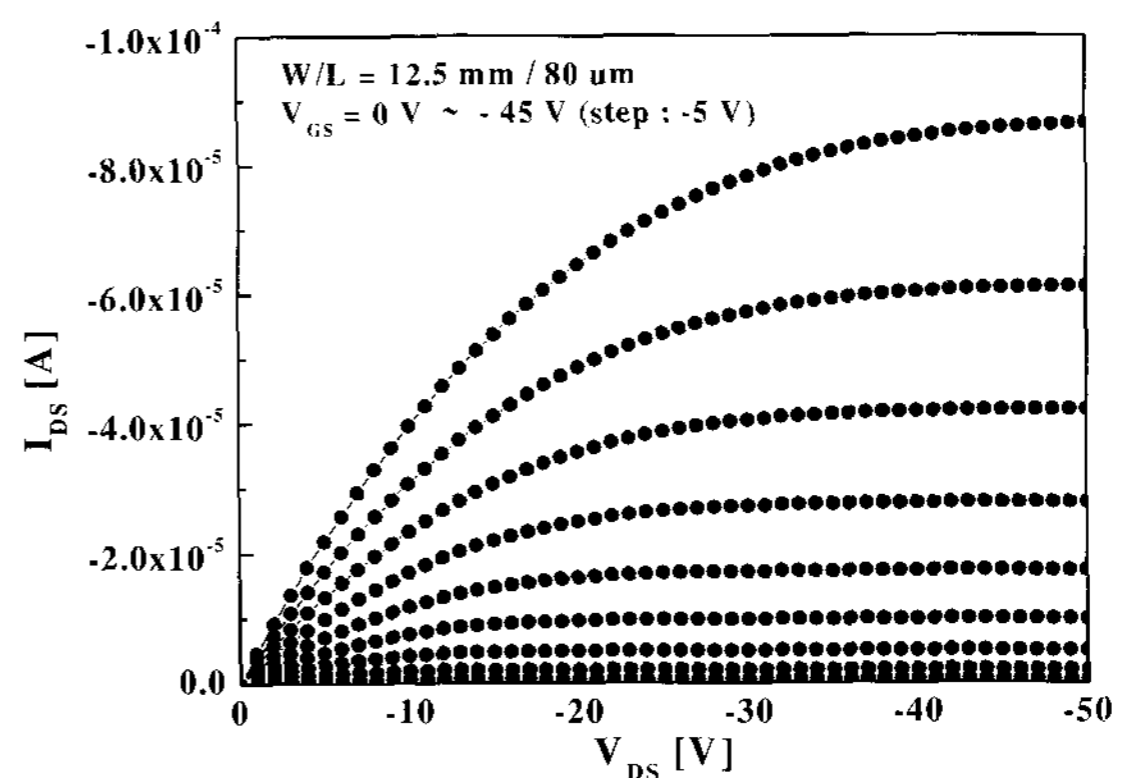


Fig. 3. The output characteristics of the individual organic thin film transistors using photoacryl as a gate insulator and pentacene as active layer.

The electrical characteristics were measured by Keithly 238 and 617 source-measurement unit. As the pentacene is a *p*-type semiconductor, and pentacene thin-film transistors operates in accumulation mode, negative

voltage was biased at the drain-source and gate-source electrodes.

3. Results and Discussion

In the paper we report of the study of driving organic electrophosphorescent devices with all-organic TFT. The characteristics of the individual organic thin film transistor using photoacryl as a gate insulator are in an enhanced mode operation as shown in Fig. 3. The drain bias (V_{DS}) was swept from 0 to -50 V and back at gate biases (V_{GS}) of -45 to 0 V at -5 V intervals. The mobility of charge carriers was 0.13 cm^2/Vs . These are typical bias conditions for p -type channel operation. It was difficult to transport the carrier in field induced channels of pentacene from one molecule to another due

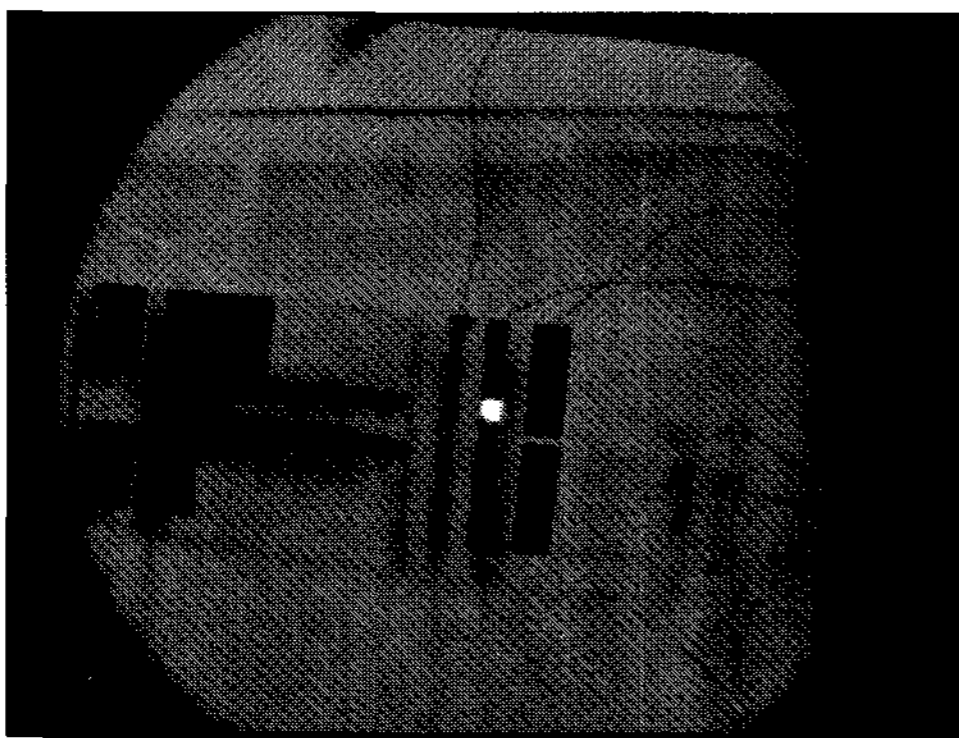


Fig. 4. Photograph of driving organic electrophosphorescent devices with all organic TFT using photoacryl as a gate insulator.

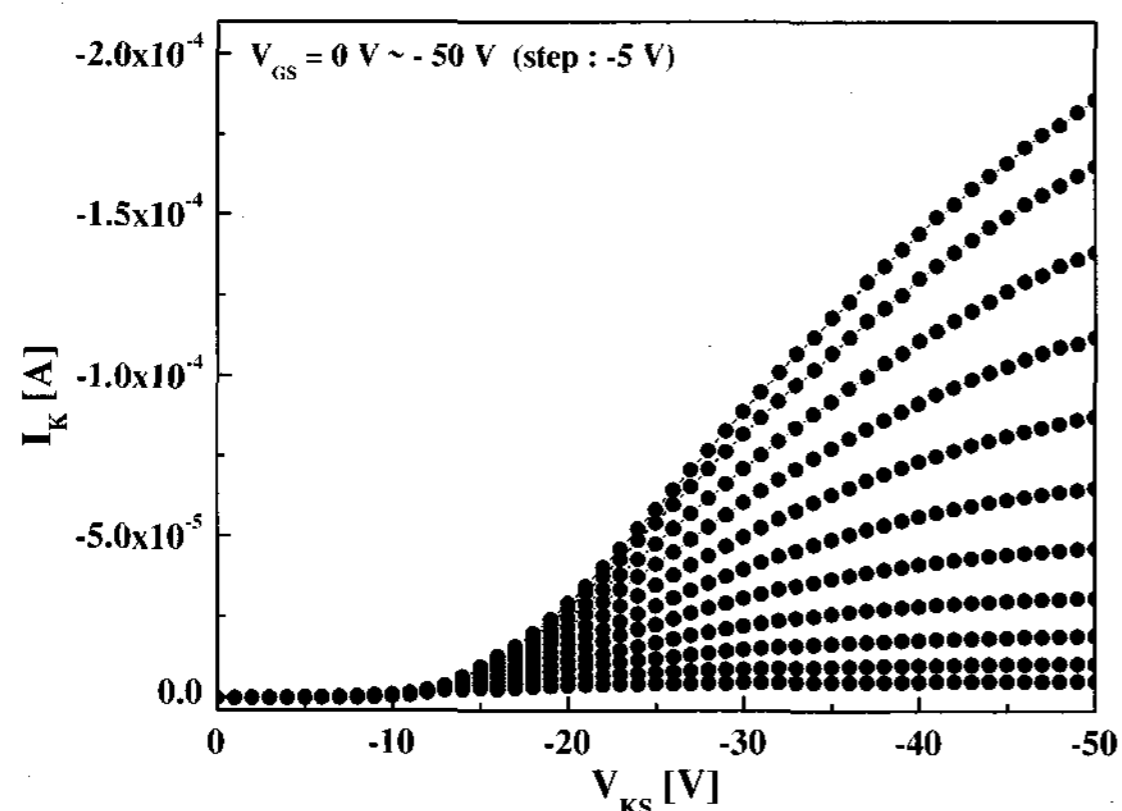


Fig. 5. The cathode current (I_K) of OLED versus the cathode-source voltages (V_{KS}) of the smart pixel of all-organic TFT with the variation of the gate-source voltage (V_{GS}). Photoacryl was used as a gate insulator.

to the disorder, defects, and chemical impurities that may from a trapping site.

We believe that the most important factor in obtaining a more satisfactory mobility as reported here in, is the improved quality of the evaporated pentacene layer.

Fig. 4 shows a photograph of driving organic electrophosphorescent devices with all-organic TFT using photoacryl as a gate insulator. Here we used silver paste for connecting device electrodes to the power supply.

Fig. 5 shows the characteristics of the cathode current (I_K) of OLED versus the cathode-source voltages (V_{KS}) where the gate-source voltage (V_{GS}) is varied. And the inset is shown simply as an electrical circuit for p -type semiconductor (in enhancement mode) [4,6] of an electrophosphorescent device with all-organic thin-film transistor using ODPA-ODA polymeric material as a gate insulator.

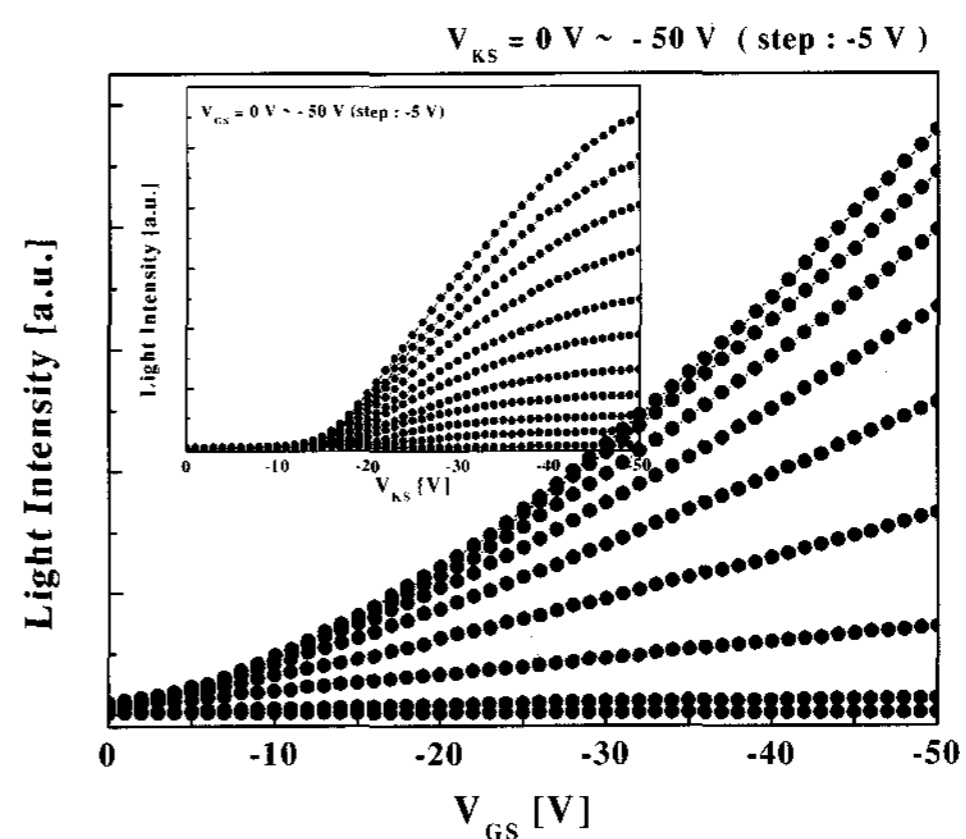


Fig. 6. The characteristics of the light intensity versus the gate-source voltages (V_{GS}) of all-organic TFT with the variation of the cathode-source voltages (V_{KS}). Photoacryl was used as a gate insulator. Inset: The characteristics of the light intensity versus the cathode-source voltages (V_{KS}) with the variation of the gate-source voltage (V_{GS}).

Fig. 6 shows the characteristics of the light intensity versus the gate-source voltage (V_{GS}) of the organic electrophosphorescent device using ODPA-ODA polymeric material as a gate insulator, where the cathode-source voltage (V_{KS}) varies. The gate bias (V_{GS}) was swept from 0 to -50 V and back at cathode-source voltages (V_{KS}) of -50 to 0 V at -5 V intervals. The inset shows the characteristics of the light intensity versus the cathode-source voltage (V_{KS}) of the organic

electrophosphorescent device where the gate voltage (V_{GS}) varies. These results show that the light intensity of organic electrophosphorescent device depends on the applied biases as cathode-source voltage (V_{KS}) and gate voltage (V_{GS}). When both V_{GS} and V_{DS} were 30 V, the maximum cathode current and the optical luminance were 31 μ A and 18.8 cd, respectively. These data extracted the maximum power luminance that was obtained about 90 cd/m^2 . Turn-on voltage was approximately 10 V. Fig. 7 shows characteristics of the luminance and the power efficiency of all-organic TFT using photoacryl as a gate insulator.

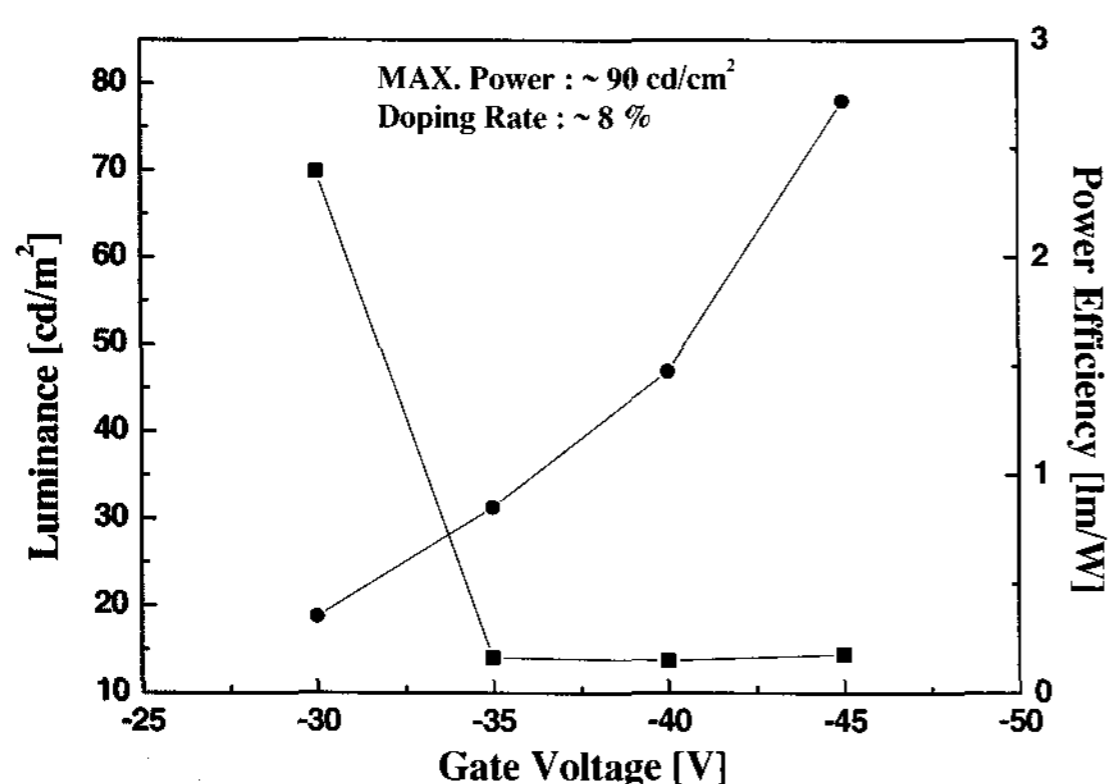


Fig. 7. The characteristics of the luminance and the power efficiency of all-organic TFT using photoacryl as a gate insulator.

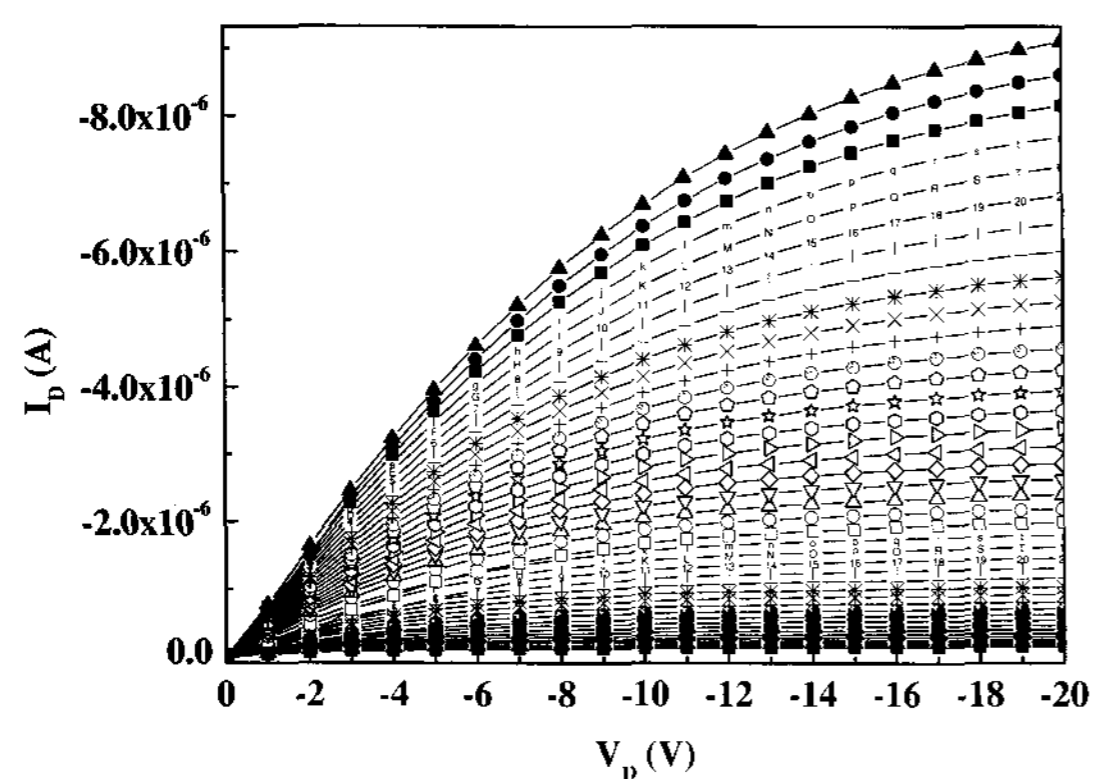


Fig. 8. Output characteristics of transistor using ODPA-ODA polymeric material as a gate insulator.

Fig. 8 shows the output I_D - V_D characteristics for an organic thin-film transistor fabricated, as described above, with the channel length and width of 50 μm and 5 mm, respectively, and a gate dielectric thickness of 0.45 μm . The field effect mobility μ_{FET} was generally

determined in the region where the drain current (I_D) saturates i.e.; μ_{FET} could be evaluated simply from $V_D > V_G - V_T$, where V_G is the gate voltage. In this region, the current can be modeled as $I_{\text{Dsat}} = (W/2L)\mu_{\text{FET}}C_i(V_G - V_T)^2$, where W and L are the channel width, and length, respectively and C_i is the capacitance of the gate dielectric layer. The field effect mobility was therefore, estimated from the slope where the data was the function of the square root of the saturation current, $I_D^{1/2}$ and of the gate voltage, V_G . As the gate voltage (V_G) and the drain voltage (V_D) increased, the drain current (I_D) also increased. Fig. 9 shows the $\log(I_D)$ - V_G characteristics for the same devices. We were able to extract a field-effect mobility of 0.134 cm^2/Vs at $V_D = -25$ V and a threshold voltage of about -7 V and. On/off current ratio was obtained at about 10^6 .

4. Conclusions

In this paper we have discussed the organic electrophosphorescent devices with all-organic TFT and have demonstrated all organic *p*-channel enhancement mode TFT using ODPA-ODA polymeric material as a gate insulator. There are many advantages of these active devices; the simplicity in circuit design and the robustness of driving circuit. The successful operation of flexible AM-OLED is necessary in order to combine phosphorescent OLED with organic TFT. We were able to demonstrate that the field effect mobility, the threshold voltage, and on-off current ratio in 0.45- μm -thick gate dielectric layer were 0.13 cm^2/Vs , -7 V, and 10^6 , respectively. Also, we were able to fabricate all organic thin film transistors using polyimide as a dielectric layer by thermal evaporation. The polyimide was doped by polymerization of ODA and ODPA at high vacuum (10^{-7} Torr), and then, cured at 150 $^\circ\text{C}$ for 1 hour. The breakdown field was found to be greater than 0.32 MV/cm.

Reference

- [1] A. Dodabalapur, Z. Bao, A. Makhija, J. G. Laquindanum, V. R. Raju, Y. Feng, H. E. Katz, and J. Rogers, *Appl. Phys. Lett.*, **73**, 142 (1998).
- [2] H. Klauk, B. D'Andrade, and T. N. Jackson, *Annual Device Research Conference Digest*, **57th**, 162 (1999).
- [3] Y. Y. Lin, D. J. Gundlach, S. F. Nelson, and T. N. Jackson,

- Jackson, IEEE Trans. Electron Devices, **44**, 1325 (1997).
- [4] D. J. Gundlach, C. C. Kuo, S. F. Nelson, And T. N. Jackson, Annual Device Research Conference Digest, **57 th**, 164 (1999).
- [5] S. W. Pyo, Y. M. Kim, J. H. Kim, J. H. Shim, and Y. K. Kim, Current Applied Physics, **2**, 417 (2002).
- [6] H. Klauk, D. J. Gundlach, J. A. Nichols, and T. N. Jackson, IEEE Trans. Electron Devices, **46**, 1258 (1999).
- [7] Yun-Myoung Kim, Sang-Woo Pyo, Jung-Soo Kim, Jae-Hoon Shim, and Young Kwan Kim, Optical Materials, **21**, 425 (2002).
- [8] D. J. Gundlach, Y. Y. Lin, T. N. Jackson, S. F. Nelson, and D. G. Schlom, IEEE Electron Device Lett., **18**, 87 (1997).