Enhanced Power Efficiency of Organic Light-Emitting Diodes using Pentacene on CF_4 -Plasma-Treated Indium Tin Oxide Anodes

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In this letter, we demonstrate high-performance organic light-emitting diodes (OLEDs) by matching various hole injection-transport layers (HITLs) with plasma surface treatments. The OLED device with pentacene HITL on CF_4-plasma-treated indium tin oxide showed the highest performance due to relatively improved hole injection and the high-hole-mobility characteristics of the pentacene. Using a high-electron-mobility material as an electron transport layer to improve the carrier balance of electrons and holes, the electroluminescent efficiency was further improved. These methods are simple and promising for the enhancement of OLED performances. These findings will be applicable to mobile displays that feature low electric power consumption.

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Young Wook Park, Hyun Ju Choi, Jin Hwan Choi, Tae Hyun Park, Jin-Wook Jeong, Eun Ho Song, and Byeong Kwon Ju, *Member, IEEE*

***Abstract*—In this letter, we demonstrate high-performance organic light-emitting diodes (OLEDs) by matching various hole injection–transport layers (HITLs) with plasma surface treatments. The OLED device with pentacene HITL on CF_4-plasma-treated indium tin oxide showed the highest performance due to relatively improved hole injection and the high-hole-mobility characteristics of the pentacene. Using a high-electron-mobility material as an electron transport layer to improve the carrier balance of electrons and holes, the electroluminescent efficiency was further improved. These methods are simple and promising for the enhancement of OLED performances. These findings will be applicable to mobile displays that feature low electric power consumption.**

***Index Terms*—Carrier mobility, CF_4-plasma (CF_4-P) treatment, hole injection–transport layer (HITL), pentacene.**

I. Introduction

SUPERIOR electroluminescent (EL) performance is a primary goal in the development of organic light-emitting diodes (OLEDs) for technological applications. Many studies have focused on surface treatments and the exploitation of organic materials for high-efficiency and long-lasting OLED device characteristics [1]–[3]. OLEDs comprising pentacene thin films have been reported previously [4]–[6]. Although pentacene has been widely used for the development of an organic thin-film transistor due to its high hole mobility (μ_h) [7], [8], has been used as the hole transport layer (HTL) of OLEDs due to its high μ_h [5], or has been used as the electron transport layer (ETL) of OLEDs due to its high electron mobility (μ_e) [6], its hole injection property in the development of high-performance OLEDs has not been systematically analyzed. In this letter, we demonstrate the potential ability of pentacene to be used as a hole injection–transport layer (HITL). To ensure a systematic analysis, as a comparison, well-known and widely used HITL materials were used, and surface treatments by various plasma treatments were applied on the indium tin oxide (ITO) anode.

Manuscript received March 14, 2011; revised May 1, 2012; accepted May 1, 2012. Date of publication June 20, 2012; date of current version July 20, 2012. This work was supported in part by the Industrial Strategic Technology Development Program (Development of Fundamental Technologies for Flexible Combined-Function Organic Electronic Device) funded by the Ministry of Knowledge Economy (Korea) under Grant KI002104, by the Basic Science Research Program through the National Research Foundation of Korea funded by the Ministry of Education, Science and Technology (MEST) under Grant CAFDC-20120000816, and by the World Class University Project by MEST under Grant R32-2008-000-10082-0. The review of this letter was arranged by Editor C. Jagadish.

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Digital Object Identifier 10.1109/LED.2012.2199461

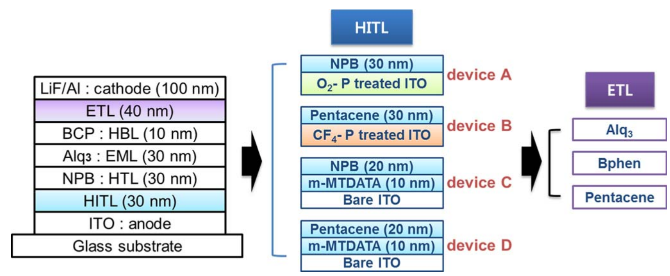

Fig. 1. Device structure of the OLEDs using four different hole injection and transport layers and three different ETLs.

Here, we demonstrate the highly improved power efficiency (PE) of OLEDs utilizing pentacene HITL on the CF_4-plasma (CF_4-P)-treated ITO (CF_4-P ITO) anode. Modification of the interface between the ITO anode and the pentacene HITL by CF_4-P treatment could improve the hole injection property and lower the driving voltage of OLEDs. When the high-μ_e material is used as an ETL, the OLEDs with pentacene HITL showed greatly improved PE due to their improved electron–hole carrier balance.

II. Device Structure

We fabricated OLEDs composed of four different types of HITLs and three different kinds of plasma treatments on the ITO anode to optimize the OLED device characteristics. (The results of the other devices will be mentioned in the supplemental section.)

Fig. 1 shows the device structure of fabricated OLEDs: 4, $4', 4''$-tris[N-(3-methylphenyl)-N-phenylamino]triphenylamine (m-MTDATA), N, N-bis(naphthalene-1-yl)-N, N-bis(phenyl) benzidine (NPB), and pentacene were used as the HITL materials; CF_4-P and O_2 plasma (O_2-P) were used for the surface treatment; device A has NPB HITL on O_2-P-treated ITO (O_2-P ITO); device B has pentacene HITL on CF_4-P ITO; devices C and D have m-MTDATA hole injection layer (HIL) on bare ITO with NPB and pentacene HTLs, respectively; and the three different ETLs for pentacene HITL on CF_4-P ITO were tris(8-hydroxy-quinolinate)aluminum (Alq_3), 4, 7-diphenyl-1, 10-phenanthroline (Bphen), and pentacene, respectively.

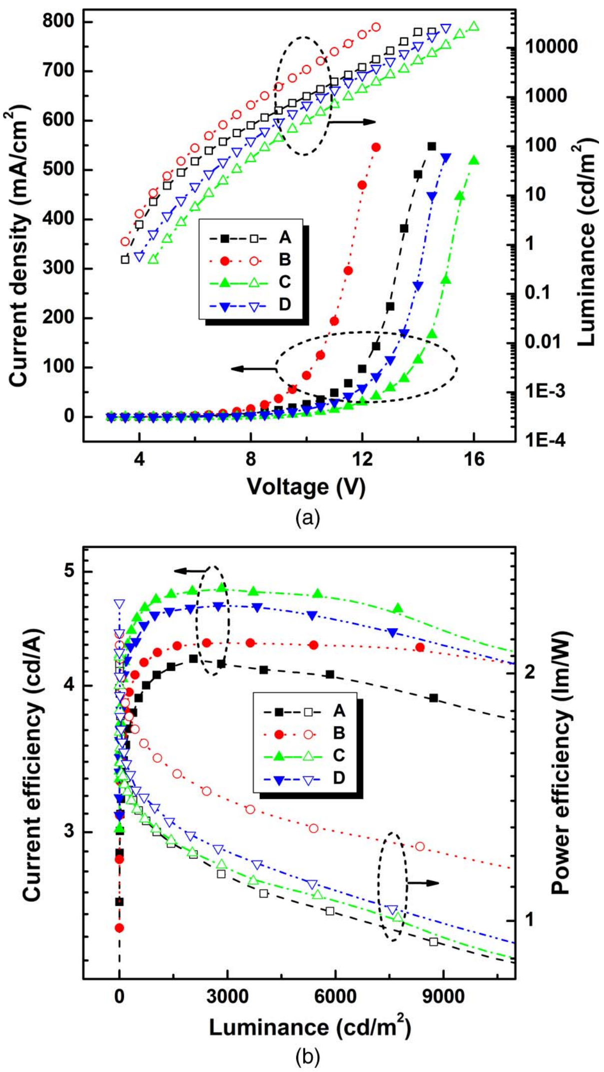

Fig. 2. (a) Current density and luminance as functions of applied voltage. (b) Current and PE as functions of luminance of the four OLED devices with different hole injection and transport layers.

III. Results and Discussion

Fig. 2(a) shows the current density and luminance as functions of the applied voltages of devices A–D. Compared to device A, device B showed much lower (∼2.5 V) driving voltage. This lower driving voltage was induced by the higher μ_h but not by the hole injection barrier. The μ_h of pentacene [8×10^{-1} $cm^2/V \cdot s$; field-effect mobility (FEM)] [7] is much lower than that of NPB [5×10^{-4} $cm^2/V \cdot s$; time of flight (TOF)] [9], while the hole injection barrier of device A is higher than that of device B: For device A, O_2-P ITO/NPB = 0.5 eV, and for device B, CF_4-P ITO/pentacene = 0 eV and pentacene/NPB = 0.6 eV. (The energy level details are described in the supplemental section.) Also, to prove the effect caused by the high μ_h of the pentacene, we compared devices C and D, which used a well-known HIL material of m-MTDATA (10 nm) as part of the HITL. Compared to device C, device D showed a lower driving voltage, even though the energy barrier at the interface of the pentacene/NPB of device D was higher than the energy barrier of the m-MTDATA/NPB of device C. Considering these results, the more efficient current-density-versus-voltage $(J{-}V)$ property of device D than device C was entirely oriented from the high mobility of the pentacene molecules. However, considering that devices C and D have the same anode–HIL structure of bare ITO/m-MTDATA and the hole injection barrier is almost the same (device C: HIL/NPB = 0.5 eV; device D: HIL/pentacene = 0 eV and pentacene/NPB = 0.6 eV), the comparable low value of the decreased driving voltage of device D compared to device C (∼1.2 V) than that of device B compared to device A (∼2.5 V) indicates that the pentacene on m-MTDATA resulted in the decrease of μ_h by the molecular orientation difference of pentacene according to the surface [7], [10], [11]. The high μ_h of pentacene on CF_4-P ITO resulted from the lower density of the interface between pentacene and CF_4-P ITO, which led to the creation of a free carrier hole [7]. In addition, the pentacene grain uniformity on the CF_4-P-treated substrate was superior to that on the bare substrate and improved the contact between the individual grains [7]. Thus, the pentacene HTL shows superior hole injection and transport characteristics compared to the conventional widely used HITL material of NPB in the optimized state (on CF_4-P ITO) and the other worse state (on m-MTDATA HIL) and resulted in the lowest driving voltage (in the order of $B < A < D < C$). (An analysis of the pentacene μ_h on plasma-treated ITO is included in the supplemental section.)

TABLE I
Summary of the PE of the OLEDs With Different HITLs

Configuration	PE at 500 cd/m^2	△PE[a]
Device A - Ref.	1.37 lm/W	100 %
Device B	1.71 lm/W	125 %
Device C	1.36 lm/W	99 %
Device D	1.44 lm/W	105 %

[a] △PE = (reference/device)

Fig. 2(b) shows the EL efficiency as a function of luminance. The PE was improved for device B compared to devices A, C, and D. As shown in Table I, the PE of device B, at 1.71 lm/W at 500 cd/m^2, was the highest. The PE of device B was elevated by 25%, 26%, and 20% compared to those of devices A, C, and D, respectively.

With regard to current efficiency (CE), in contrast to the tendency of the PE, device B was not prominent over the measured luminance range. The lower CE of device B compared to those of devices C and D was caused by an electron–hole carrier imbalance induced by the increased excess holes of the pentacene HITL. In this electron-injection-limited device, formation of the exciton could shift due to the increased excess holes, leading to a declined CE. The CE of pentacene-HITL-based OLEDs would be improved by the application of high ETL materials such as Bphen [12].

Fig. 3 shows the EL efficiency ratio of the OLEDs with different ETLs. On the contrary to the previous result using pentacene as the ETL in inverted OLEDs [6], the EL properties of these devices are not in order of μ_e; rather, they are ordered in Bphen ($\mu_e = 10^{-4}$ $cm^2/V \cdot s$; TOF) [12] > Alq_3 ($\mu_e = 10^{-6}$ $cm^2/V \cdot s$; TOF) [14] > pentacene ($\mu_e = 5 \times 10^{-3}$ $cm^2/V \cdot s$; FEM) [13]. This result occurs due to unbalanced electron–hole mobility. Compared to the carrier mobility of the Alq_3 ($\mu_h = 10^{-8}$ $cm^2/V \cdot s$ and $\mu_e = 10^{-6}$ $cm^2/V \cdot s$) [14], the pentacene has an $\sim 10^7$ higher μ_h and an10^3 higher μ_e. Although pentacene has the highest μ_e among the three

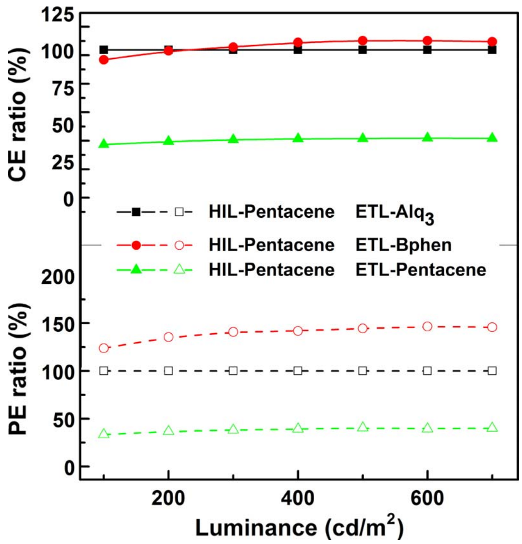

Fig. 3. EL efficiency ratio and luminance of the three OLEDs (the reference device with Alq_3 ETL is a standard of 100%.) (CE) Current efficiency. (PE) Power efficiency. (HIL) Hole injection layer. (ETL) Electron transport layer.

materials and can be used as an ETL in the inverted OLEDs at a specified system [6], the device using the pentacene HITL and ETL showed the lowest CE due to a poor carrier balance caused by unbalanced electron–hole mobility. However, Bphen has a 10^2 higher μ_e, a much lower μ_h than Alq_3 (the μ_h of Bphen is too low to be measured [12]), and the highest efficiency. The Bphen ETL device shows the highest CE ratio of 106%. Controlling the carrier mobility, the device has an efficient electron–hole balance, which leads to an optimized exciton formation. This is the reason for the improved CE of the OLED device using the Bphen ETL.

The PE ratio shows a much higher dependence than the CE ratio. The device using the Bphen ETL showed a highly improved PE ratio of 144%. In conclusion, the OLED device with the pentacene HITL on CF_4-P ITO and the Bphen ETL showed balanced electron–hole mobility and a resultant highly improved PE.

IV. Conclusion

OLEDs with a pentacene HITL on the CF_4-P ITO showed a PE (1.71 lm/W) that was even higher than those of the widely used HITLs of the O_2-P ITO/NPB (1.37 lm/W) and the m-MTDATA (1.44 lm/W). Using the high-μ_e material of Bphen as the ETL, the PE was highly improved by 44% due to improved electron–hole carrier balance using balanced carrier mobility. The high μ_h of the pentacene with the CF_4-P surface treatment resulted in improved carrier injection and transport of OLEDs. This leads to improved PE, low operating voltage, and reduced electric power consumption for the OLED devices.

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