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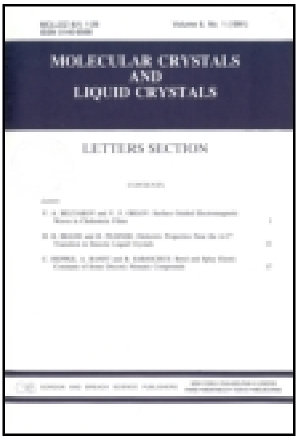

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Ho-Nyeon Lee [a] & Mun Soo Choi [b]

[a] Department of Display and Electronic Information Engineering , Soonchunhyang University , Asan , 336-745 , Korea

[b] Department of Electric & Robotics Engineering , Soonchunhyang University , Asan , 336-745 , Korea

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Dependence of the Characteristics of Photovoltaic Organic Light-Emitting Diodes on Device Structure

HO-NYEON LEE[1],* AND MUN SOO CHOI[2]

[1]Department of Display and Electronic Information Engineering,
Soonchunhyang University, Asan 336-745, Korea
[2]Department of Electric & Robotics Engineering, Soonchunhyang University,
Asan 336-745, Korea

Photovoltaic organic light-emitting diodes (PVOLEDs) that have dual functions of organic light-emitting diodes and organic solar cells have been studied. Using the structure of glass/ITO anode/hole transporting layer (HTL)/5,6,11,12-tetraphenylnaphthacene (rubrene)/C_{60}/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)/LiF/Al, we examined the dependence of properties for PVOLEDs on HTL materials. Using N,N'-bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) with high hole mobility and high lowest-unoccupied molecular orbital (LUMO) level as a HTL material, we obtained the PVOLED that operated as an organic solar cell with 1.86% power conversion efficiency under reverse bias and as an organic light-emitting diode under forward bias with approximately 700 cd/m^2 luminance at 7 V.

Keywords Organic light-emitting diode; organic solar cell; photovoltaic organic light-emitting diode

Introduction

There is growing interest on organic electronic devices such as organic light-emitting diodes (OLEDs) and organic solar cells (OSCs) due to the success within the commercial market of OLEDs. Currently, OLEDs are mostly used for mobile devices such as smart phones and smart pads. OLED panel and glass substrate sizes have to increase largely in order to widen their application range to large-size televisions. In addition, reduction of production costs, increase of lifetime and improvement of light-emission efficiency are required. Silicon based solar cells have been the major photovoltaic device until now. This is because the performance of solar cells using other materials is inferior to that of silicon based solar cells. Researches on OSCs are focused on the increase of efficiency and lifetime to a commercial product level. Flexible OSC is acquiring attention because of its ability to increase the application range of OSCs. OLEDs and OSCs have similar diode structures fabricated using organic semiconducting materials. There have been attempts to combine both the light-emitting function and the photovoltaic function in a device. This photovoltaic organic light-emitting diode (PVOLED) must be useful in expanding the application area of organic electronics. PVOLEDs can be used for various new applications such as extremely

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*Address correspondence to Prof. Ho-Nyeon Lee, Department of Display and Electronic Information Engineering, Soonchunhyang University, 646, Eupnae-ri, Shinchang-myeon, Asan 336-745, Korea (ROK). Tel.:(+82)41-530-4703; Fax: (+82)41-530-1548. E-mail: hnlee@sch.ac.kr high-efficient displays, outdoor stand-alone digital information displays, and photovoltaic smart windows.

Electrons and holes have to recombine and generate light quickly in order to make efficient OLEDs. However, electrons and holes must separate quickly to make efficient OSCs. The device structures of OLEDs and OSCs are optimized for electron hole recombination and electron hole separation, respectively. Therefore, PVOLEDs having the dual functions are difficult to be realized. There have been a few reports about dual function organic diodes. Several device structures were used for PVOLEDs such as conventional organic diode structures [1–5], ionic junctions [6], half-gap junction structures [7] and tandem structures [8]. Previous reports have demonstrated the feasibility of PVOLEDs. However, the efficiencies of electric power generation and light emission were much lower than those of conventional OLEDs and OSCs. The PVOLEDs with the conventional organic diode structures show photovoltaic behavior under not white light but UV light. The ionic junction PVOLEDs uses ionic materials, which have possibility to cause the stability issue due to the migration of the ions. The tandem PVOLEDs are composed of two units of a light-emitting unit and a photovoltaic unit, so the stacking structure is complex. Using a structure of indium tin oxide (ITO) anode/poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS)/5,6,11,12-tetraphenylna phthacene (rubrene)/C_{60}/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)/metal cathode, the half-gap junction PVOLEDs showed the 2.9% power conversion efficiency (PCE) under air mass (AM) 1.5 illumination.

In this work, we have studied the dependence of PVOLED performance on hole transporting layers (HTLs). For various HTLs, we examined the device characteristics of PVOLEDs with half gap junction structures.

Experiments

PVOLEDs were fabricated using 0.5 mm thick glass substrates. Plasma beam deposited ITO with a sheet resistance of 10 Ω/□ was used as an anode electrode. Anodes were patterned using a photolithography and a wet etch method. After the anode patterning, oxygen-plasma treatment was conducted using inductively coupled plasma equipment. Organic layers and a cathode layer were then deposited sequentially using a thermal evaporation method at a pressure of $10–^{6}$ Torr. After the fabrication of diode structures, an encapsulation process was conducted using a glass cap in a nitrogen environment. After the encapsulation process, PVOLED characteristics were measured. Light-emitting properties were measured using a spectroradiometer in a dark box. The photovoltaic properties were measured using a solar simulator at the AM 1.5 condition.

In this work, we used devices with rubrene/C_{60} junctions. Rubrene acts as a light-emitting layer when the device operates as an OLED and as an electron-donating layer when the device operates as an OSC. C_{60} acts as an electron-accepting layer when the device operates as an OSC. The device structure except for HTL, was fixed in this work. The stack structure of the device was a glass substrate/ITO/HTL/rubrene /C_{60}/BCP/LiF/Al. ITO, BCP, LiF and Al were used as an anode, electron transporting layer, electron injection layer and a cathode, respectively. The thicknesses of ITO, rubrene, C_{60}, LiF and Al layers were 150 nm, 45 nm, 35 nm, 1 nm and 100 nm, respectively. As HTL layers, phthalocyanine (CuPC), 4,4',4''-tris(2-naphthylphenyl-phenylamino)triphenylamine (2-TNATA) and N,N'-bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) were tested. The thicknesses of CuPC, 2-TNATA and NPB were 20 nm, 15 nm, and 30 nm, respectively.

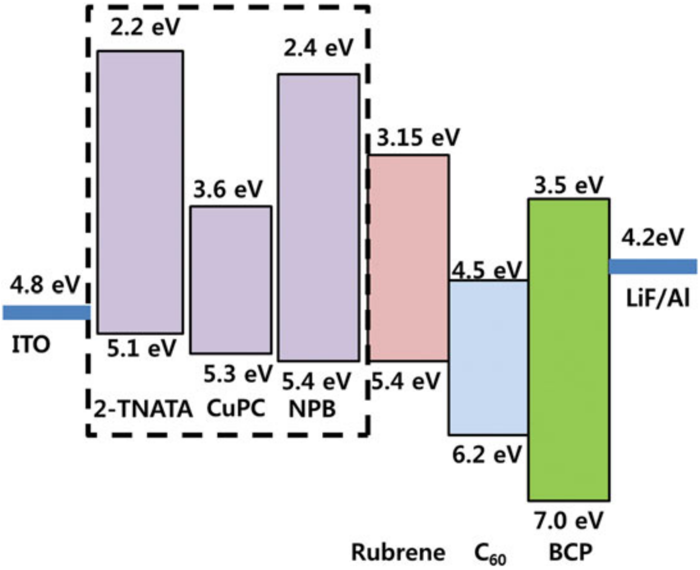

Figure 1. Schematic diagram of the energy bands for PVOLEDs of this work.

The energy band diagram of PVOLEDs for this work is shown in Fig. 1. The box with a dashed line means HTL materials; one of them was selectively used as a HTL layer for a PVOLED device. As shown in the figure, the highest occupied molecular orbital (HOMO) levels of HTL materials were in the range between the Fermi level of ITO and the HOMO level of rubrene.

Results and Discussion

Current density as a function of bias voltage between the anode and cathode is shown in Fig. 2(a). 2-TNATA HTL decreased the current density considerably. Current densities of PVOLEDs with NPB HTL and with CuPC HTL were similar with each other. PVOLED without HTL had the highest current density. As shown in Fig. 1, the HOMO levels of 2-TNATA, CuPC and NPB are all similar with one another. Hole mobilities of 2-TNATA, CuPC and NPB are 6.3×10^{-5} $cm^2V^{-1}s^{-1}$[9], $10^{-3}\sim10^{-4}$ $cm^2V^{-1}s^{-1}$[10] and $(8.8\pm2)\times10–^{4}$ $cm^2V^{-1}s^{-1}$[11], respectively. Therefore, the smallest current density of PVOLED with 2-TNATA HTL may be due to the lowest hole mobility of 2-TNATA among the HTL materials of this work. The largest current density of PVOLED without HTL means that the injection barrier between ITO and rubrene is not critical for PVOLED with regards to the structure of this work. Luminance as a function of bias voltage is shown in Fig. 2(b). As shown in Fig. 1, the difference between the HOMO levels of rubrene and C_{60} is not negligible. Therefore, direct injection of the electron from C_{60} to rubrene is difficult to achieve. As shown in Fig. 1, the HOMO level of C_{60} is approximately located at the middle level of the HOMO and LUMO of rubrene. Electrons at the LUMO level of C_{60} can recombine with holes at the HOMO level of rubrene. Other electrons at the LUMO level of C_{60} can absorb the released energy (LUMO of C_{60} – HOMO of rubrene). These electrons can be stimulated to the LUMO level of rubrene due to the absorbed energy. Excitons in rubrene can be formed through these procedures. Therefore, rubrene emits light despite the large difference between the LUMO levels of C_{60} and rubrene. PVOLEDs with CuPC HTL

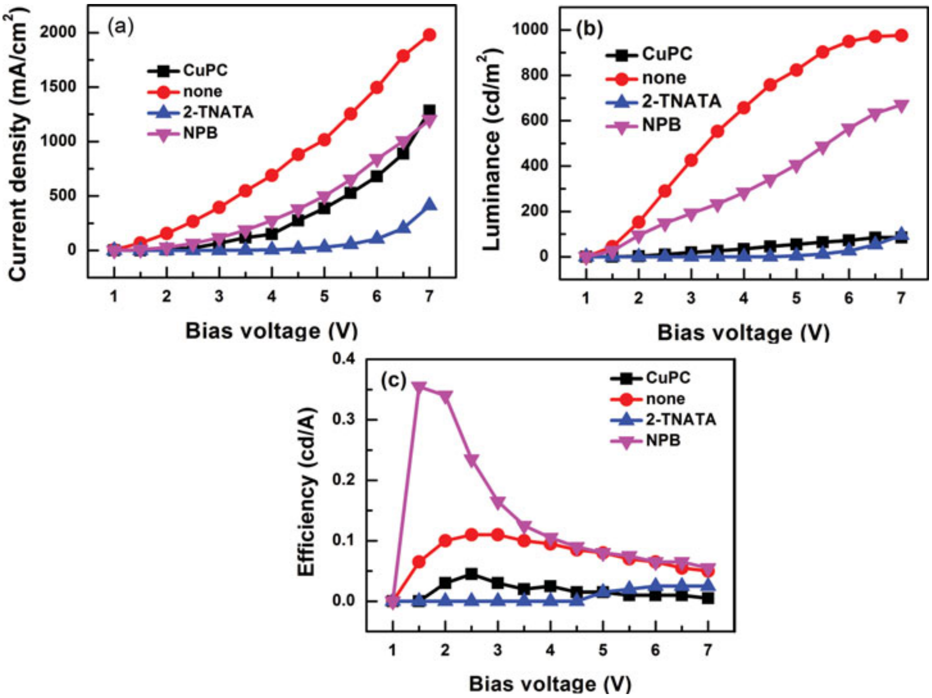

Figure 2. (a) Current density, (b) luminance and (c) current efficiency of PVOLEDs as a function of forward bias voltage.

and 2-TNATA HTL had much low luminance compared with other PVOLEDs. PVOLED with 2-TNATA HTL had low current density, so its luminance had to be low. PVOLED with CuPC HTL did not have a low current density; its low luminance may be explained by the low LUMO level of CuPC as shown in Fig. 1. Electrons in rubrene can go through to CuPC instead of participating in the recombination process because the LUMO level of

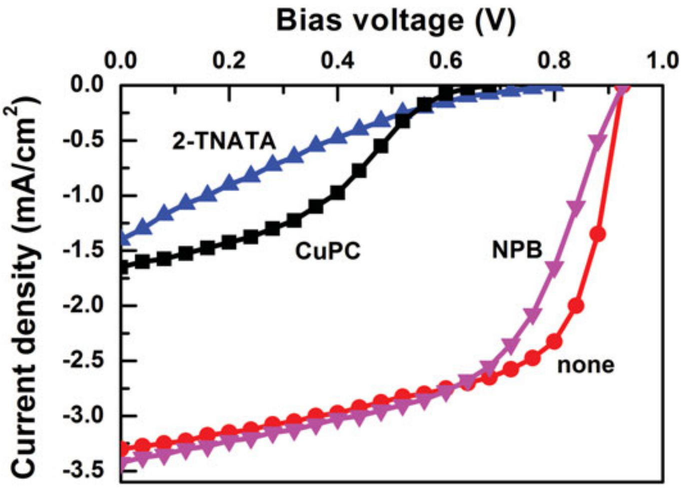

Figure 3. Current density of PVOLEDs as a function of reverse bias voltage.

Table 1. Properties of PVOLEDs as OSCs according to HTL

HTL	PCE (%)	FF (%)	V_{OC} (V)	J_{SC} (mA/cm^2)
None	1.89	62.05	0.92	3.31
NPB	1.86	52.98	0.93	3.79

CuPC is lower than that of rubrene. The highest luminance of PVOLED without HTL is related with that its current density is highest among PVOLEDs of this work. Efficiency as a function of bias voltage is shown in Fig. 2(c). PVOLEDs with 2-TNATA HTL and CuPC HTL had low efficiency. PVOLED with NPB HTL had higher efficiency compared to PVOLED without HTL. The lower hole current of PVOLED with NPB HTL may cause a better charge balance between holes and electrons; the improved charge balance increases the efficiency.

Current density as a function of reverse bias voltage regarding PVOLEDs is shown in Fig. 3. This property represents the function of PVOLED as an OSC; the illumination condition was AM 1.5. As shown in the figure, PVOLEDs with CuPC HTL and 2-TNATA HTL had weak properties compared to PVOLEDs with NPB HTL and without HTL. Short circuit current density (J_{SC}) and open circuit voltage (V_{OC}) of PVOLED with NPB HTL were higher than those of PVOLED without HTL. However, fill factor (FF) of PVOLED with NPB HTL was slightly smaller than that of PVOLED without HTL. The additional series resistance due to the NPB layer may cause this lower FF. The device parameters as OSCs of PVOLEDs with NPB HTL and without HTL are summarized in Table 1.

Conclusions

Using a rubrene/C_{60} junction structure, we successfully fabricated PVOLEDs, which could operate as OLED or OSC according to the bias conditions. The dependence of PVOLED characteristics on HTL materials was studied using structures of ITO/HTL/rubrene/C_{60}/BCP/LiF/Al. 2-TNATA was not appropriate as a HTL material for this structure because of its low hole mobility. Also, CuPC was not proper as a HTL material for this structure because of its low LUMO level. PVOLED with NPB HTL had good properties as an OLED and as an OSC; the relatively high hole mobility of NPB and high LUMO level may cause these strong properties. From the PVOLED with NPB HTL, we could obtain a 1.86% PCE, 52.98% FF, 0.93 V V_{OC} and 3.79 mA/cm^2 J_{SC}. Luminance and current efficiency of this PVOLED with NPB HTL was approximately 700 cd/m^2 at 7 V bias and about 0.35 cd/A.

Acknowledgments

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