

Enhanced Electroluminescence Efficiency of Phosphorescent Organic Light-Emitting Diodes by Controlling the Triplet Energy of the Hole-Blocking Layer

Young Wook Park, Young Min Kim, Jin Hwan Choi, Tae Hyun Park, Jin-Wook Jeong, Hyun Ju Choi, and Byeong Kwon Ju, *Member, IEEE*

Abstract—This letter reports on the effect of the triplet energy (E_T) of the hole-blocking layer (HBL) on triplet exciton quenching between the emissive layer (EML) host and the HBL of phosphorescent organic light-emitting diodes (PHOLEDs). Using different EML hosts and HBLs having different E_T 's and electron mobilities, the effects of the E_T 's of the HBL have been analyzed. When the E_T of the HBL is lower than that of the EML host, the PHOLEDs show significant dependence of reduced device performances. PHOLEDs having a 3-(4-biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole HBL show 65% improved external quantum efficiency (EQE) at 500 cd/m² with a 4,4',4''-tris(*N*-carbazolyl)-triphenyl-amine host than that with an *N,N'*-dicarbazolyl-4-4'-biphenyl host, while PHOLEDs having a 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline HBL show 41% reduced EQE. To solve the remaining key issue of developing highly efficient PHOLEDs, the E_T of matching the EML and the HBL is extremely desirable and is also explored.

Index Terms—Phosphorescent organic light-emitting diodes (PHOLEDs), triplet energy of hole-blocking layer (HBL), triplet quenching.

I. INTRODUCTION

PHOSPHORESCENT organic light-emitting diodes (PHOLEDs) have been widely investigated recently because of their high potential to realize perfect 100% internal quantum efficiencies [1], [2]. Recent research based on efficient triplet exciton confinement and reducing the triplet exciton quenching has reported many accomplishments [3], [4]. These results are based on controlling the triplet excitons in the emissive layer (EML) because they are the main emissive substance in PHOLEDs. From these results, it is possible

Manuscript received October 6, 2009; revised January 19, 2010. Date of publication March 15, 2010; date of current version April 23, 2010. This work was supported in part by the Korean Science and Engineering Foundation under Grant R11-2007-045-01003-0 funded by the Korean Ministry of Education, Science and Technology (MEST), by World Class University Project R32-2008-000-10082-0 of MEST, and by the IT R&D program of MKE/IITA (2009-F-017-01, Development of Fundamental Technologies for Flexible Combined-Function Organic Electronic Device). The review of this letter was arranged by Editor C. Jagadish.

The authors are with the Display and Nanosystem Laboratory, College of Engineering, Korea University, Seoul 136-713, Korea (e-mail: zerook@korea.ac.kr; kimjang@korea.ac.kr; jinncm@korea.ac.kr; thpark@korea.ac.kr; ilovejin@korea.ac.kr; guswn1020@korea.ac.kr; bkju@korea.ac.kr).

Color versions of one or more of the figures in this letter are available online at <http://ieeexplore.ieee.org>.

Digital Object Identifier 10.1109/LED.2010.2041891

TABLE I
ENERGY LEVELS AND ELECTRON MOBILITY OF ORGANIC MATERIALS

Materials	HOMO, LUMO (eV)	E_T^a (eV)	Electron mobility (cm ² /Vs, at 0.5MV/cm)
TCTA	-5.7, -2.3 [3]	2.92 [12]	
mCP	-5.9, -2.4 [4]	2.98 [7]	
CBP	-6.1, -3.0 [5]	2.64 [6]	
BCP	-6.5, -3.0 [8]	2.58 [6]	5.5E-6 [10]
TAZ	-6.6, -2.6 [9]	>>2.76 ^b	1.0E-6 [11]
TPBi	-6.3, -2.9 [10]	2.70 [12]	3.3E-5 [10]

^aTriplet energy taken from the onset wavelength of the phosphorescence spectrum

^bIn this work, the triplet energy was calculated as $3.26\text{eV} \geq E_T >> 2.76\text{eV}$; see the supplemental for further details.

to presume that reducing the triplet exciton quenching by triplet-energy (E_T) matching of the EML and the HBL or the HTL (depending on the location of the emission zone) is most important in achieving highly efficient PHOLEDs. Although Lee *et al.* [4] have reported that the electron mobility of the HBL is more important than its E_T , it is not clear whether the dependence of the E_T of the HBL on the triplet exciton quenching at the EML/HBL interface is significant or not because all HBLs have much lower E_T 's than the EMLs in their device configurations.

This letter investigates the relationship between the E_T of the HBL and the triplet exciton quenching at the EML/HBL interface. To make a systematic analysis, different EML hosts and HBLs with different E_T 's are prepared and studied in detail.

II. DEVICE STRUCTURE

The energy levels, including the highest occupied molecular orbital (HOMO), the lowest unoccupied molecular orbital (LUMO), and the E_T 's of various organic materials, are presented in Table I.

The device structures of the two batches of PHOLED devices, where each batch has three subdevices employing different HBLs, are shown in Fig. 1. The main difference between the two batches is the emissive host materials, namely, the CBP ($E_T = 2.64$ eV) and the TCTA ($E_T = 2.92$ eV). In addition, to prevent the triplet exciton quenching at the HTL/EML interface,

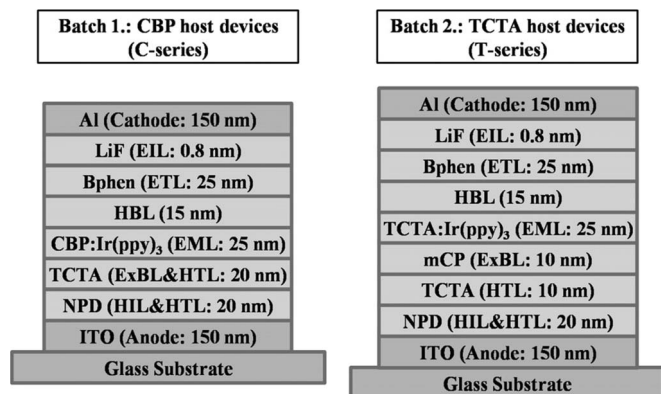


Fig. 1. Device structures of the fabricated PHOLEDs.

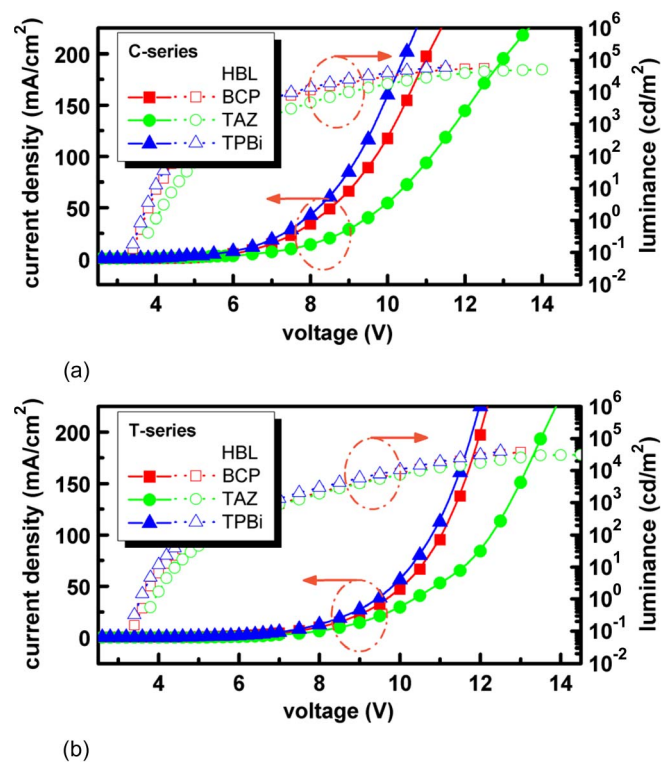


Fig. 2. Current density and luminance as a function of applied voltage for (a) C-series and (b) T-series devices.

different exciton-blocking layers (ExBL) of the TCTA and the mCP ($E_T = 2.98$ eV) for batch-1 and batch-2 devices, respectively, have been applied to confine the triplet excitons into the EML.

III. RESULTS AND DISCUSSION

Figs. 2 and 3 show the electroluminescence (EL) characteristics of batch-1 devices (CBP host devices, C-series) and batch-2 devices (TCTA host devices, T-series). The summarized performances of devices are presented in Table II.

The overall EL characteristics depend on the HBL material. From Fig. 2(a) and (b), in both of the C-series and T-series devices, the driving voltages are increased on the order of TPBi < BCP < TAZ, in the same order of the electron mobilities and the hole-injection barrier at the EML/HBL interface. Because

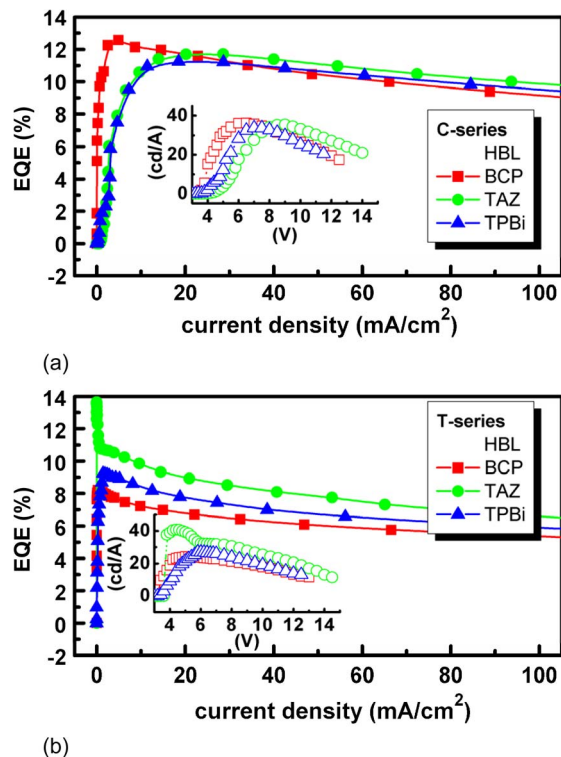


Fig. 3. EQE as a function of current density for (a) C-series and (b) T-series devices. The inset shows current efficiency as a function of applied voltage.

 TABLE II
 PERFORMANCE OF THE FABRICATED PHOLEDs BY HBLs

Efficiency	BCP		TAZ		TPBi	
	cd/A	EQE(%)	cd/A	EQE(%)	cd/A	EQE(%)
C-series	peak	36.2, 12.6	35.2, 11.7	33.7, 11.3		
	at 500cd/m ²	31.9, 10.6	17.7, 6.0	17.6, 5.9		
T-series	peak	24.1, 8.2	40.8, 13.6	26.9, 9.2		
	at 500cd/m ²	23.3, 7.9	32.1, 10.7	26.9, 9.2		
Δ EQE ^a	peak	-35%	+16%	-19%		
	at 500cd/m ²	-41%	+65%	+33%		

$$^a \Delta \text{EQE} = (\text{T-series/C-series} - 1)$$

the devices with a TAZ HBL have the highest carrier-injection barriers at both of the hole-injection barrier of the EML/HBL interface and the electron-injection barrier of the HBL/ETL interface, the TAZ HBL has the highest driving voltage. From the aforementioned result, it is seen that the carrier-injection barrier determines the overall driving voltage of the fabricated OLEDs. Comparing the C- and the T-series, the overall driving voltages are increased in the T-series. These increased driving voltages come from the increased electron- and hole-injection barriers at the TCTA host/HBL interface ($\Delta \text{LUMO}_{\text{TCTA-CBP}} = 0.7$ eV, $\Delta \text{HOMO}_{\text{TCTA-CBP}} = 0.3$ eV).

In Fig. 3(a), among the C-series devices, the overall external quantum efficiency (EQE) at the high-current-density (> 20 mA/cm²) region are on the order of TAZ (3.26 eV $\geq E_T \gg 2.76$ eV) > TPBi ($E_T = 2.70$ eV) > BCP ($E_T = 2.58$ eV), which suggests that they follow the order of the E_T , not the order of the electron mobility or the hole-injection barrier at the EML/HBL interface. In contrast, PHOLEDs with a BCP HBL show the highest EQE at low current densities

(< 20 mA/cm²). These EL characteristics of the low-current-density region depend more on the carrier-injection property, i.e., the lowest electron-injection barrier of BCP at the HBL/ETL interface results in the highest EQE at low current densities.

In Fig. 3(b), the T-series shows a clearer dependence on HBLs; the EQEs are on the order of the E_T that is the same as for the C-series, but the differences between HBLs are much bigger. However, the EQE of the T-series devices (BCP and TPBi HBL) decrease more than that of the C-series devices, except that the TAZ HBL devices show improved characteristics in lower current densities (< 9 mA/cm²). The BCP HBL device shows the greatest reduced peak EQE of -35%, while that of the TPBi and TAZ devices are -19% and +16%, respectively. The BCP and TPBi devices have lower E_T than the TCTA device. Thus, triplet quenching between the TCTA host and the HBL is increased, and as a consequence, the performances of the BCP and TPBi HBL devices in the T-series are reduced. On the other hand, the TAZ has a higher E_T , which is enough to confine the excitons into the EML, than the other HBLs, thereby preventing triplet quenching between the EML host and the HBL. As a result, the T-series TAZ HBL device shows the highest improved performances in the lower current density region (< 9 mA/cm²) among all of the C- and T-series devices.

Because the TAZ HBL devices of the T-series show the most improved EL efficiencies at lower current densities (< 9 mA/cm²), the overall EQEs of all the T-series devices (BCP, TAZ, and TPBi) in the high-current-density region (> 9 mA/cm²) are reduced. In addition, compared to the C-series devices, the T-series devices show higher efficiency roll-off that arises from the lower recombination rate resulting from the disappeared electron-injection barrier at the ExBL/EML interface in the T-series [4]. Because the mCP ExBL has a slightly lower LUMO than the TCTA host, the electrons could easily penetrate from the TCTA into the ExBL without generating the excitons in the EML. Despite this efficiency roll-off of the T-series devices, the effect of the E_T level of the HBL is clearly shown by the order of the EL efficiency.

IV. CONCLUSION

When the E_T of the HBL is lower than that of the EML host, triplet quenching becomes the dominant nonemissive quenching process. Considering the E_T of organic materials: comparing the CBP host devices with the TCTA host devices, the TCTA host devices with BCP and TPBi HBLs show significantly reduced peak EQEs (-35%, -19% reduced), while

that of the TAZ HBL devices show a +16% improved EQE. In particular, a luminance of 500 cd/m² for the TAZ HBL device with the TCTA host shows the highest improved EQE (+65%) than that with the CBP host device, while the BCP HBL device shows significantly reduced EQE (-41%) with the TCTA host. These results suggest that the exciton-blocking property of the HBL should be considered in developing highly efficient PHOLEDs in order to prevent triplet quenching. The matching of the E_T of not only the HTL/EML interface but also the EML/HBL interface is an important key issue in developing highly efficient PHOLEDs.

REFERENCES

- [1] M. A. Baldo, D. F. O'Brien, Y. You, A. Shoustikov, S. Sibley, M. E. Thompson, and S. R. Forrest, "Highly efficient phosphorescent emission from organic electroluminescent devices," *Nature*, vol. 395, no. 6698, pp. 151-154, Sep. 1998.
- [2] M. A. Baldo, S. Lamansky, P. E. Burrows, M. E. Thompson, and S. R. Forrest, "Very high-efficiency green organic light-emitting devices based on electrophosphorescence," *Appl. Phys. Lett.*, vol. 75, no. 1, pp. 4-6, Jul. 1999.
- [3] F. X. Zang, T. C. Sum, A. C. H. Huan, T. L. Li, W. L. Li, and F. Zhu, "Reduced efficiency roll-off in phosphorescent organic light emitting diodes at ultrahigh current densities by suppression of triplet-polaron quenching," *Appl. Phys. Lett.*, vol. 93, no. 2, pp. 023 309-1-023 309-3, Jul. 2008.
- [4] J. Lee, N. Chopra, S.-H. Eom, Y. Zheng, J. Xue, F. So, and J. Shi, "Effects of triplet energies and transporting properties of carrier transporting materials on blue phosphorescent organic light emitting devices," *Appl. Phys. Lett.*, vol. 93, no. 12, pp. 123 306-1-123 306-3, Sep. 2008.
- [5] H. Kanno, Y. Sun, and S. R. Forrest, "High-efficiency top-emissive white-light-emitting organic electrophosphorescent devices," *Appl. Phys. Lett.*, vol. 86, no. 26, pp. 263 502-1-263 502-3, Jun. 2005.
- [6] I. Tanaka and S. Tokito, *Highly Efficient OLEDs With Phosphorescent Materials*, H. Yersin, Ed. Weinheim, Germany: Wiley-VCH Verlag GmbH & Co. KgaA, 2008.
- [7] R. J. Holmes, S. R. Forrest, Y. J. Tung, R. C. Kwong, J. J. Brown, S. Garon, and M. E. Thompson, "Blue organic electrophosphorescence using exothermic host-guest energy transfer," *Appl. Phys. Lett.*, vol. 82, no. 15, pp. 2422-2424, Apr. 2003.
- [8] R. J. Holmes, S. R. Forrest, T. Sajoto, A. Tamayo, P. I. Djurovich, M. E. Thompson, J. Brooks, Y. J. Tung, B. W. D'Andrade, M. S. Weaver, R. C. Kwong, and J. J. Brown, "Saturated deep blue organic electrophosphorescence using a fluorine-free emitter," *Appl. Phys. Lett.*, vol. 87, no. 24, pp. 243 07-1-243 507-3, Dec. 2005.
- [9] C. Adachi, M. A. Baldo, M. E. Thompson, and S. R. Forrest, "Nearly 100% internal phosphorescence efficiency in an organic light-emitting device," *J. Appl. Phys.*, vol. 90, no. 10, pp. 5048-5051, Nov. 2001.
- [10] Y. Q. Li, M. K. Fung, Z. Xie, S.-T. Lee, L.-S. Hung, and J. Shi, "An efficient pure blue organic light-emitting device with low driving voltages," *Adv. Mater.*, vol. 14, no. 18, pp. 1317-1321, 2002.
- [11] H. Sasabe, E. Gonmori, T. Chiba, Y.-J. Li, D. Tanaka, S.-J. Su, T. Takeda, Y.-J. Pu, K.-I. Nakayama, and J. Kido, "Wide-energy-gap electron-transport materials containing 3,5-dipyridylphenyl moieties for an ultra high efficiency blue organic light-emitting device," *Chem. Mater.*, vol. 20, no. 19, pp. 5951-5953, Sep. 2008.
- [12] S. Reineke, F. Lindner, G. Schwartz, N. Seidler, K. Walzer, B. Lussem, and K. Leo, "White organic light-emitting diodes with fluorescent tube efficiency," *Nature*, vol. 459, no. 7244, pp. 234-238, May 2009.