

RESEARCH ARTICLE | SEPTEMBER 07 2005

The effect of interfacial layer on the performance of organic light-emitting diodes ✓

Stelios A. Choulis; Vi-En Choong; Mathew K. Mathai; Franky So

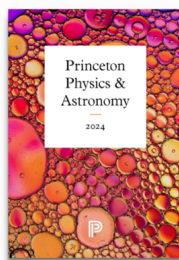


Appl. Phys. Lett. 87, 113503 (2005)

<https://doi.org/10.1063/1.2042635>



05 June 2024 15:45:25



Browse our new Physics and Astronomy Catalog
30% off titles with code **P326**

 PRINCETON UNIVERSITY PRESS

The effect of interfacial layer on the performance of organic light-emitting diodes

Stelios A. Choulis,^{a)} Vi-En Choong, Mathew K. Mathai, and Franky So
Osram Opto Semiconductors Inc, San Jose, California 95134

(Received 16 May 2005; accepted 20 July 2005; published online 7 September 2005)

We demonstrate high efficiency organic light-emitting devices by incorporation of a nanoscale interfacial layer between a hole-conducting layer (PEDOT:PSS) and a light-emitting polymer layer (LEP) to improve hole injection. The interfacial layer has appropriate highest occupied molecular orbital level in order to act as a bridge for efficient hole injection from the PEDOT:PSS into the LEP. As an example we have incorporated a suitable interfacial layer into a green-emitting single-layer electrophosphorescent light-emitting diode. Devices with the interfacial layer show a peak efficiency of 41 lm/W, an improvement of more than 25% in their performance over comparable devices without the interfacial layer. The results presented here introduce a novel method to improve hole injection and thus efficiency in organic electroluminescent devices. © 2005 American Institute of Physics. [DOI: 10.1063/1.2042635]

Organic light-emitting diodes (OLEDs), attract considerable interest from the research community due to their applications in flat-panel displays and solid state lighting.¹ The emission properties of OLEDs active region can be divided in two broad categories: fluorescent and phosphorescent emitters. In particular, phosphorescent emitters open the possibility to develop highly efficient OLEDs since they are able to achieve emission from both singlet and triplet excited states.² Although solution processed OLEDs offer the potential for low fabrication cost, easy processing, and flexibility, it is more challenging to achieve good charge balance.³ Incorporation of small molecules (molecular doping) into the fluorescence and phosphorescence emissive region or synthesis of novel polymers has been used to improve the charge injection and transport.^{4–7}

In terms of electrophosphorescence-based OLEDs, the triplet energy levels of the materials involved is of crucial importance since luminescence quenching effects are likely to occur and deteriorate the device efficiency.^{6,8} The interplay between quenching and injection/transport properties determines their efficiency. High-band-gap host materials with higher triplet energy than that of the phosphorescent dye are usually used to minimize quenching. In particular, poly(N-vinylcarbazole) (PVK) polymer has been shown to insure the confinement of the triplet excited state in the guest molecules and thus prevent luminescence quenching.⁹ Recently, Yang *et al.*⁵ have developed a single-layer highly efficient electrophosphorescent OLED by using tris(2-(4-tolyl)phenylpyridine)iridium Ir(mppy)₃ doped into a PVK polymer host. The PVK: Ir(mppy)₃ matrix was co-doped with 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) and N,N'-diphenyl-N-N'-(bis(3-methylphenyl)-[1,1-biphenyl]-4-4'-diamine (TPD) as the electron- and hole-transporting moieties, respectively. Devices with power conversion efficiency up to 24 lum/watt at luminous efficiencies exceeding 30 cd/A were reported.⁵ In this letter we report improvement on the performance of a similar electrophosphorescence OLEDs by incorporation of an interfacial layer between the hole-conducting (buffer) layer and the light-emitting layer to improve hole injection prop-

erties. We have substantially improved the efficiency of electrophosphorescent OLEDs by assisting direct hole injection to phosphorescent dye and eliminating/reducing additional quenching effects usually observed by incorporation of low triplet energy hole-injecting layers into device architecture.

The Ir(mppy)₃ phosphorescence emitter, Poly(9,9-dioctylfluorene) (PFO) and poly(9,9-dioctyl-fluorene-co-N-(4-butylphenyl)-diphenylamine) (TFB) used for interfacial layers were purchased from American Dye Sources. Poly(3,4-ethylenedioxy thiophene) doped with poly(styrene sulfonate) (PEDOT:PSS) used for buffer layer was purchased from H. C. Starck. PVK, TPD, and PBD were purchased from Sigma-Aldrich and used without any further purification.

The light-emitting layer of the devices is a blend of PVK, TPD, PBD, and Ir(mppy)₃ sandwiched between two injecting electrodes. The anode consists of indium tin oxide (ITO)/glass substrates precoated with a PEDOT:PSS buffer layer. Prior to spin coating with the PEDOT:PSS buffer layer, ITO was precleaned and UV ozone treated. After the UV ozone treated step, a 60 nm PEDOT:PSS layer was spin coated onto the ITO. The PEDOT:PSS layer was baked in nitrogen environment at 200 °C for 30 min. Blends of PVK, PBD, and Ir(mppy)₃ in chlorobenzene solutions were spin coated on top of PEDOT:PSS layer inside the glove box with conditions to yield a light-emitting layer thickness of around 70 nm. The light-emitting layer was then annealed at 80 °C for 30 min.⁵ Following the spin coating step, the samples were transferred into the thermal evaporator in nitrogen atmosphere. Devices were then completed by thermal evaporation of a shadow masked, CsF (1 nm) and aluminum (200 nm) top contact, at a typical pressure of 10⁻⁶ mbar. Following evaporation, the devices were encapsulated with a glass lid and getter.

In the present study, TFB and PFO were used as interfacial layers between PEDOT:PSS and light-emitting layer. The interlayer was spin coated inside the glove box onto the PEDOT:PSS buffer layer from toluene solution at 2000 rpm/60 s and baked at 130 °C for 10 min in a nitrogen environment. Under these conditions an interlayer thickness of around 10 nm was obtained. To achieve a nanometer scale interfacial layer and prevent mixing of the interlayer with the

^{a)}Electronic mail: stelios.choulis@osram-os.com

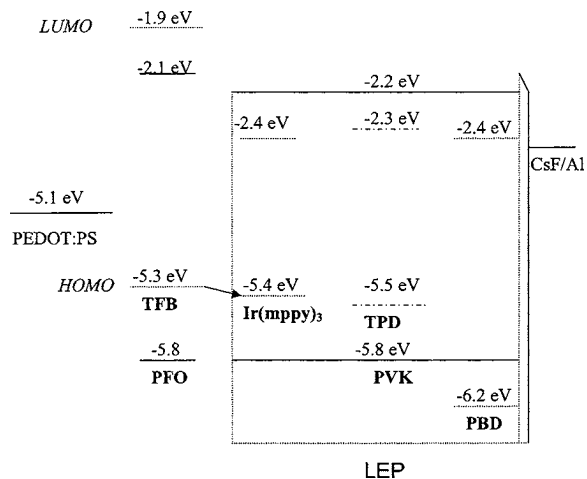


FIG. 1. The HOMO and lowest unoccupied molecular orbitals energy levels of TFB and PFO used for interfacial layers and Ir(mppy)₃, PVK, TPD, and PBD components of the LEP compared to the work function of PEDOT:PSS (all referenced with respect to vacuum). The vector in the scheme indicates the direct injection from the TFB interfacial layer into Ir(mppy)₃ phosphorescent organometallic compound within the LEP.

light-emitting layer, we introduced a toluene wash step: a neat toluene was dispensed onto the interlayer followed by spinning at 1000 rpm for 30 s. This produces an interfacial layer with thickness of a few nanometers for both cases.¹⁰

The device architectures studied are described as follows. The control device is based on ITO/PEDOT:PSS/light-emitting polymer layer(LEP)/CsF/Al. To investigate the effect of interfacial layer in the hole injection properties of the device, we have incorporated a TFB or PFO interfacial layer between the PEDOT:PSS and the LEP layer (ITO/PEDOT:PSS/interfacial TFB or PFO/LEP/CsF/Al). Similar to previously reported data, the LEP contains [61% PVK + 24% PBD + 9% TPD + 6% Ir(mppy)₃].⁵ The concentrations are given by weight.

TFB is particularly suitable for interfacial layer in green electrophosphorescence devices since its HOMO level is aligned with the highest occupied molecular orbital (HOMO) level of TFB and Ir(mppy)₃ at around 5.4 eV,^{5,11} as shown in Fig. 1. In contrast, the HOMO level of PFO interlayer is 5.8 eV,¹¹ significantly lower than the HOMO level of Ir(mppy)₃ and very similar to PVK,⁵ as also shown in Fig. 1. We discuss this point further later in the text.

Figure 2 shows the current density/voltage (J/V) properties (upper plot) and the brightness-voltage properties (lower plot) of the devices under study. From the J/V characteristics, it is clear that the current density is much larger for the device incorporating an interfacial layer of TFB compared to the control device. In J/V measurements, the increase in the current density can be related to both injection and bulk transport properties. However the ultrathin TFB interfacial layer used here is very unlikely to affect the bulk transport properties. As we have also discussed previously, the preparation of the interfacial layers indicates that no mixing with the light-emitting layer has occurred. Furthermore, the devices were not exposed to air, so that additional doping effects can be ruled out. Our results indicate that the improvement in the current density is related to improved hole injection by incorporating a TFB interfacial layer into the device architecture. The importance of selecting material with the appropriate HOMO level for interfacial layer is indicated by the incorporation of the PFO interfacial layer into

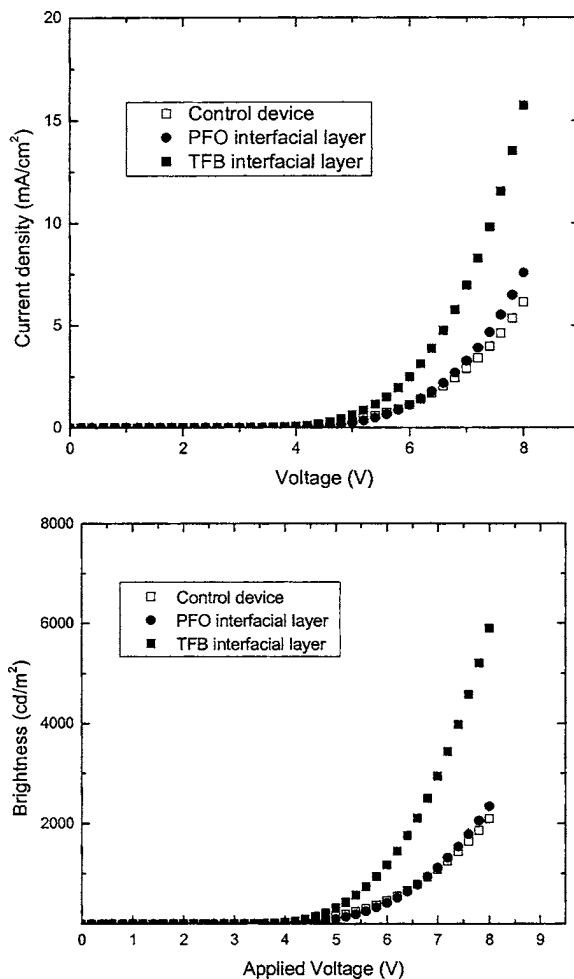


FIG. 2. Upper plot: Current density-voltage characteristics. Lower plot: Brightness-voltage characteristics of the OLEDs under study. The control device is represented with open squares, the devices incorporating a TFB and PFO interfacial layer between PEDOT:PSS represented with solid squares and solid circles respectively.

the device architecture. In this case, the large hole injection barrier from PEDOT:PSS into the LEP is not changed and no improvement in the current density compared to the control device is obtained.

Further proof for the improvement in hole injection by incorporation of TFB interfacial layer can be seen in the brightness-voltage properties of the devices shown in the lower plot of Fig. 2. The brightness of the device incorporating TFB as an interfacial layer is higher through the whole voltage range compared to the control device, indicating a lower driving voltage for devices with an interfacial TFB layer. This is another indication that the TFB interfacial layer improves hole injection properties. In contrast, no improvement in brightness/voltage characteristics is shown for devices using interfacial PFO layer. Further, one may consider that incorporation of interfacial layer (with the process conditions described previously) is to condition the PEDOT:PSS surface, providing a more intimate interface between PEDOT:PSS and LEP. An improvement in the interface between PEDOT:PSS and LEP should also improve hole injection. However, the clear difference between the incorporation of PFO and TFB interfacial layers rules out any possibility for the incorporated interfacial layer to just improve the interface between PEDOT:PSS and LEP layer. The increase in device current and brightness observed with the interfacial

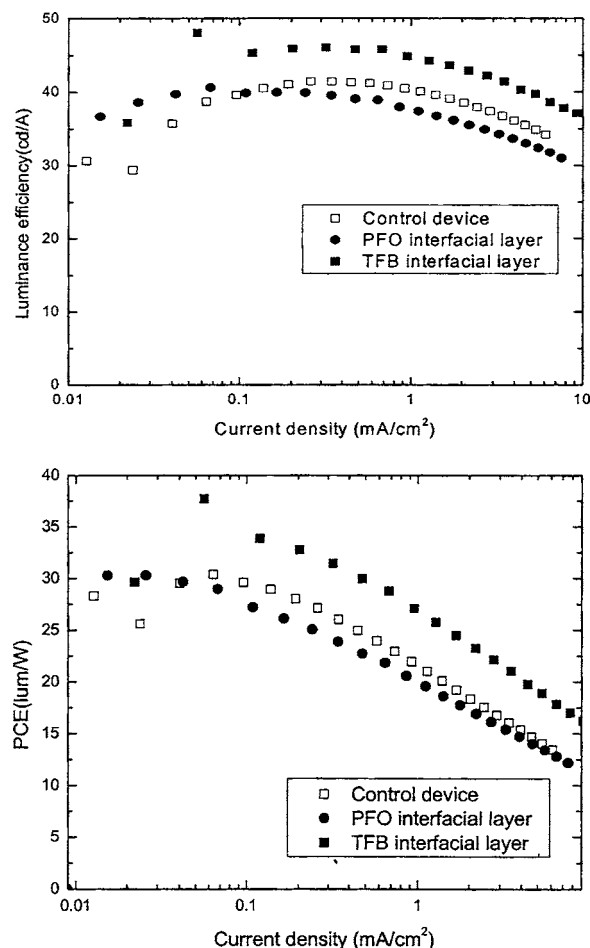


FIG. 3. Upper plot: Luminance efficiency-current density characteristics. Lower plot: Power conversion efficiency-voltage characteristics of the OLEDs under study. The control device is represented with open squares, the devices incorporating a TFB and PFO interfacial layer between PEDOT:PSS represented with solid squares and solid circles, respectively.

TFB layer is attributed to the matching of HOMO levels for hole injection from the interfacial layer into the LEP.

Figure 3 upper plot shows the luminescence efficiency versus current density properties and the power conversion efficiency versus current density properties (lower plot) for the devices under study. The peak power conversion efficiency (PCE) of the control device was 30 lm/watt with a luminance efficiency reaching 40 cd/A. The performance of the devices using PFO interfacial was similar to that of the control devices. The PFO interfacial layer has a HOMO level similar to the PVK host and does not aid in hole injection.

Finally, devices with TFB interfacial layer show the highest PCE of 38 lm/W at luminescence efficiencies exceeding 45 cd/A. The result just presented is based on devices prepared in the same experimental series, using the same batch of materials, the same solutions and preparation conditions in order to ensure direct comparison for the data presented. The PCE achieved by incorporating TFB interfacial layer into device architecture can be as high as 41 lm/W at luminescence efficiencies reaching 50 cd/A. This indicates that incorporation of the TFB interfacial layer can be used to achieve high efficiency OLED devices.

Due to their similar HOMO levels, the TFB interfacial layers act as a bridge for efficient hole injection into Ir(mppy)₃ dye and TPD. The present results indicate that direct injection into the HOMO level of the phosphorescent

organometallic compound is of high importance to obtain an efficient electrophosphorescence device. It should be noted here that the triplet energy levels of PFO and TFB are in the range of 2.2–2.3 eV,¹² much lower than the triplet energy level estimated from the peak photoluminescence emission maximum of Ir(mppy)₃ at 2.44 eV. Thus, one might expect that quenching effects are very likely to occur.¹³ The strong improvement on the device performance by the incorporation of TFB interfacial layer is due to improved hole injection, which results in the moving of the recombination zone away from the anode. In this case, any quenching due to the lower triplet energy level of TFB is minimized. However, in the case of PFO, no improvement of hole injection is seen. Thus, one may expect to observe lowered device performance due to quenching effects by PFO. The similar device performance between the control device and devices incorporating PFO interfacial layer indicates that the nanoscale interface modification introduced here does not result in additional luminescence quenching effects usually observed in phosphorescent OLEDs by incorporation of hole-transporting/injecting layers with low triplet energy excited state.⁸

In summary incorporation of interfacial layer (with a HOMO energy level similar to that of the phosphorescence dye used within the LEP) between the PEDOT:PSS hole-transporting layer and LEP can be used to assist direct hole injection to the phosphorescence compound without introducing additional strong luminance quenching effects, independent of the triplet energy levels of the materials involved. By introducing TFB interfacial layer into Ir(mppy)₃/PVK/TPD/PBD solution processed OLED, the driving voltage was reduced and a high PCE was achieved. The same technique applies to fluorescence-based OLEDs, where the interfacial layer should be chosen such that the HOMO energy level of the interfacial layer lies between the HOMO energy level of the light emitting layer and the work function of the anode.¹⁴

This work is supported by the Department of Energy (Polymer OLED White Light Development Program; award number DE-FC26-04NT41947).

¹D. D. C. Bradley, *Curr. Opin. Solid State Mater. Sci.* **1**, 789 (1996).

²M. A. Baldo, D. F. O'Brien, Y. You, A. Shoustikov, S. Sibley, M. E. Thompson, and S. R. Forrest, *Nature (London)* **395**, 151 (1998).

³S. A. Choulis, Y. Kim, J. Nelson, D. D. C. Bradley, M. Giles, M. Shkunov, and I. McCulloch, *Appl. Phys. Lett.* **85**, 3890 (2004).

⁴G. E. Johnson, K. M. McGrane, and M. Stolka, *Pure Appl. Chem.* **67**, 175 (1995).

⁵X. H. Yang and D. Neher, *Appl. Phys. Lett.* **84**, 2476 (2004).

⁶M. Suzuki, S. Tokito, F. Sato, T. Igarashi, K. Kondo, T. Koyama, and T. Yamaguchi, *Appl. Phys. Lett.* **86**, 103507 (2005).

⁷B. W. D'Andrade and S. R. Forrest, *Adv. Mater. (Weinheim, Ger.)* **16**, 1585 (2004).

⁸Y. Wang, *Appl. Phys. Lett.* **85**, 4848 (2004).

⁹J. Pina, J. S. de Melo, H. D. Burrows, A. P. Monkman, and S. Navaratnam, *Chem. Phys. Lett.* **400**, 441 (2004).

¹⁰The term interfacial layer is used to describe an ultrathin layer between PEDOT:PSS and LEP. Due to the method followed to prepare the interfacial layers (involving a wash step as discussed in the text), accurate determination of the actual thickness of such a thin layer is not possible using the conventional method of profilometry.

¹¹M. Redecker, D. D. C. Bradley, M. Inbasekaran, W. W. Wu, and E. P. Woo, *Adv. Mater. (Weinheim, Ger.)* **11**, 241 (1999).

¹²D. Hertel, Y. V. Romanovskii, B. Schweitzer, U. Scherf, and H. Bassler, *Macromol. Symp.* **175**, 141 (2001).

¹³M. Sudhakar, P. I. Djurovich, T. E. Hogen-Esch, and M. E. Thompson, *J. Am. Chem. Soc.* **125**, 7796 (2003).

¹⁴S. A. Choulis, V. Choong, A. Patwardhan, M. K. Mathai, and F. So (unpublished).