



Low reflection and photo-sensitive organic light-emitting device with perylene diimide and double-metal structure

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ABSTRACT

In this paper, an organic light-emitting device (OLED) with low reflection for the entire visible range and at different viewing angles by using an absorptive and photo-sensitive material, N,N'-Bis(2,6-diisopropylphenyl)-1,7-bis(4-methoxyphenyl)perylene-3,4,9,10-tetracarboxydiimide (MPPDI), as a black layer (BL) structure was demonstrated. A semitransparent double-metal structure, Al/Ag, was used to reduce the driving voltage and enhance the destructive interference of the BL structure. Combined with the broad absorption band of the MPPDI, the reflectance of such a device shows a 12-times decrease compared to the conventional OLED. Also, a photocurrent enhancement effect was observed in our BL-OLED due to the photosensitive quality of the MPPDI.

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1. Introduction

It is advantageous to use organic light-emitting device (OLED) due to its light weight, self emission, wide view-angle, high luminance efficiency, low drive voltage, and fast response time, all of which make OLED one of the most promising display technologies for the next generation [1,2]. However, ambient reflection from the metal electrode of OLED deteriorates the ambient contrast ratio (A-CR) and limits its applications in bright environment, especially for outdoor displays [3–5]. There are many approaches to rectify this drawback such as the use of a polarizer or a filter laminated on the OLED panel, or the insertion as a light-absorptive layer and/or a destructive interference layer into the OLED. The latter is called the “black layer” (BL) technology, which mainly reduces reflection from the metal cathode, without extra cost for the laminating process [6–16]. The following reported reflectances of devices with different absorptive structures inside OLED are indicated for entire visible spectrum.

The light-absorptive layer with high conductivity, such as an Al layer doped with the oxygen monoxide (relative reflection of device ~5%–12%) [6], and a mixture of metal with the organic material (sun/eye-integrated reflectance of device ~9%–12%) [7,8], can be inserted between the electron transport layer (ETL) and the cathode electrode. Besides, the conductive electrode could be an absorptive layer itself (for example, stacked cathodes embodied with aluminum oxide nano-

clusters (reflectance of device ~12%–0.16%) [9], or mixing absorptive Sm with Ag by co-evaporation (reflectance of device >15%) [10]). In these cases, absorption determines the reflectance of devices and therefore a highly absorptive material for the whole visible spectrum (a “black” material) is required.

For the BL-OLEDs with a destructive interference effect, the highly reflective electrode is usually replaced by the stacked “thin metal-organic-thick metal” structure (reflectance of device ~5%–12%) [11–14]. The reflected light from the semi-transparent thin metal and the thick metal interferes destructively with a suitable cavity design. The selection criteria for the organic materials include high thermal stability and high carrier mobility, with candidates tris-(8-hydroxyquinoline) aluminum (Alq₃) [11–13]. Further, the organic layer is absorptive itself and further helps to enhance the A-CR effectively [14]. Generally, it is not easy to achieve low reflection for the whole visible spectrum and at different view angles without absorption mechanism, because the cavity design of the destructive interference is dependent on the wavelength and light-incident angle. Since the emission from the OLED toward the cathode has been absorbed, the maximum efficiency of a BL device is typically one-half of the conventional one, whether absorptive or destructive [14]. Those photons absorbed by the BL relax their energy by heat, which is an energy waste, and possibly shorten the operation lifetime [17].

In this paper, the authors present a BL-OLED combining the absorption and destructive interference effect. The absorptive material is N,N'-Bis(2,6-diisopropylphenyl)-1,7-bis(4-methoxyphenyl)perylene-3,4,9,10-tetracarboxydiimide (MPPDI) [18–20], whose chemical structure is shown in the inset of Fig. 1. Its lowest unoccupied

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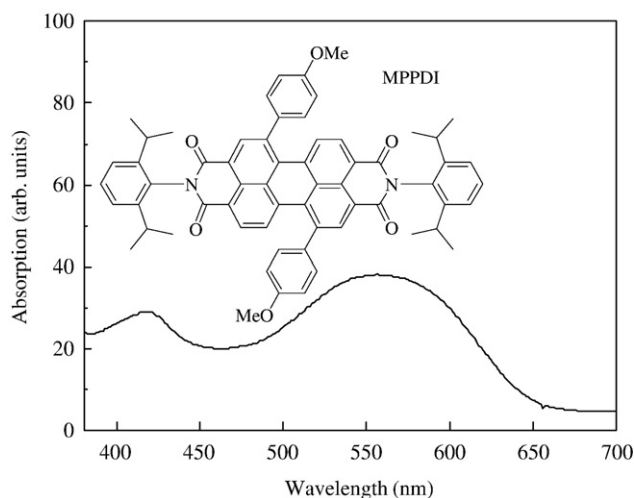


Fig. 1. Absorption spectrum and chemical structure of MPPDI. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) are 3.97 eV and 5.9 eV, respectively. MPPDI is a perylene derivative which exhibits high electron mobility and is commonly used for organic photovoltaic applications [21]. By suitable molecular design, this material has two absorption bands at blue (418 nm) and yellowish-green (557 nm) as shown in Fig. 1. The two bands were caused by the absorption of the perylene core, and by the photoinduced intramolecular charge-transfer effect [18], which features charge transfer from aryl substituents to the perylene diimide core due to photon energy absorption, respectively. To achieve an effective destructive interference, a semi-transparent double-metal-layer, Al/Ag, was used to optimize the proportion of transmittance and reflectance to decrease the spectral reflectance from 418 to 557 nm. Hence, a BL-OLED with low reflectance for the whole visible range can be achieved. Notably, the perylene materials are widely applied to organic photovoltaic devices due to their good electron transport characteristics and high absorption coefficient for visible range. In our BL-OLED, we observed that it was sensitive to the ambient light and accompanied a photo-induced current inside the BL part. Such a photo-induced current could separate out electrons and holes by a good structural design, OLEDs integrated on solar cell (reflectance of device ~1.4%–12% with anti-reflection coating) [22], which has the functions applied for power-saving, ambient light detection, and self-brightness-tuning.

2. Experiment

Our OLED fabrication and basic characterization details are outlined in Ref. [14]. The spectroradiometer, Konica Minolta CS1000, and digital source-meter, Keithley 2400, were configured for measur-

ing electroluminescence intensity and spectrum, current density (J) and driving voltage (V) of devices. The spectrophotometer, Hitachi U-4100, was used to measure reflectance of devices from ultraviolet to near infra-red at different wide viewing angles, and A-CR could then be calculated by the following equation [10]:

$$A - CR = \frac{\text{Device luminance (On)} + \text{Reflected ambient light}}{\text{Device luminance (Off)} + \text{Reflected ambient light}} \quad (1)$$

For studying the photo-current effect of our devices, J-V characteristics under the excitation of a 532 nm green laser were measured by a Keithley 2400 source-meter.

Eleven samples were fabricated with their device structures shown in Table 1. In these devices, we used N, N-Bis (naphthalen-1-yl) -N, N-bis (phenyl) benzidine (NPB) as the hole-transport layer, Alq₃ as the emitting layer and the ETL, LiF as the electron injection layer and Al/Ag/MPPDI/LiF/Al as the BL cathode. The semi-transparent metal consisted of a single Al (Ag) layer or a double metal Al/Ag, which provides efficient destructive interference and an electron injection pathway resulting from the large LUMO difference between Alq₃ and MPPDI. Between the MPPDI and the Al cathode, an electron injection layer LiF was applied out of necessity for better electron injection from the cathode into MPPDI.

Devices 1 to 4 were designed to study the electron injection and transport characteristics of MPPDI. Workfunctions of electrodes, Ag and Al, are 4.26 eV and 4.28 eV, respectively. Those values are close to the LUMO value of the MPPDI, and far from the HOMO value of the MPPDI. Thus, only the electron current could exist in these four devices. Device 5 was a conventional highly reflective bottom-emitting OLED. Devices 6 to 9 had different semi-transparent layer structures, and devices 7, 10 and 11 were designed for different MPPDI thicknesses.

3. Results and discussion

3.1. Electrical characteristics

Fig. 2a shows the J-V curves of electron-only devices 1 to 4, where Ag is used as an anode. Fixing the bias at 4 V, the current densities of devices 1 to 4 are 8.27×10^{-6} , 0.0874, 0.341, and 0.0416 A/cm², respectively. The current density of MPPDI (device 3) is 8.2-times higher than that of Alq₃ (device 4), which clearly shows the superior electron transport characteristics of MPPDI. By comparing the J-V characteristics of device 2 and 4, we can see that the “intrinsic” carrier transport characteristic of MPPDI is better than that of Alq₃. Devices 1 and 3 are designed to study the electron injection characteristics from the electrode into the organic MPPDI layer. A thin LiF layer following the Al electrode (device 3) shows a higher current density than only the Al electrode itself (device 1), which may result from the Li-ion doping effect [23] or a sharp density of states coupling effect of the thin LiF layer [24]. Fig. 2b shows the J-V curves of the control device (device 5) and low reflection devices (devices 6 to 11). The thin metal

Table 1
Layer structures of the OLED devices.

| | Anode | NPB | Alq ₃ | LiF | Al | Ag | MPPDI | Alq ₃ | LiF | Cathode | | |
|-----------|-------|--------|------------------|-----|------|----|-------|------------------|-----|---------|-----|---|
| Device 01 | } | Ag 200 | } | } | } | } | 500 | } | } | } | | |
| Device 02 | | | | | | | 500 | | | | 20 | 5 |
| Device 03 | | | | | | | 500 | | | | 5 | |
| Device 04 | | | | | | | 520 | | | | 5 | |
| Device 05 | } | } | } | } | 1500 | } | } | } | } | | | |
| Device 06 | | | | | 50 | | | | | | | |
| Device 07 | | | | | 25 | | | | | 25 | 750 | |
| Device 08 | | | | | 10 | | | | | 40 | 750 | |
| Device 09 | | | | | 50 | | | | | 750 | | |
| Device 10 | | | | | 25 | | | | | 25 | 650 | |
| Device 11 | 25 | 25 | 850 | | | | | | | | | |

Unit:Å.

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